Response to anonymous Referee #1

We sincerely thank the reviewers for their constructive and thoughtful suggestions, which improve the quality of this paper. We have made the revisions and responses following your comments point by point.

The referee comments are shown in black.
The responses to the comments are shown in blue. The line numbers refer to the clean version of our revised manuscript.
The changes included in the revised manuscript are shown in red.

1 General comments

General comment 1:
This paper developed a calculation framework (APFoam-1.0), based on open-source CFD code OpenFOAM, for atmospheric photolysis to examine the micro-scale reactive pollutant formation and dispersion in the urban area. Five new types of reaction are added to the chemistry module, which is coupled with full O3–NOx–VOCs chemistry and CFD model. The model was validated against SAPRC box modeling software and wind tunnel experimental data. The framework was applied to case studies investigating O3–NOx–VOCs formation processes and dispersion of the reactive pollutants in an example of a typical street canyon. APFoam provides a numerical simulation tool based on the general purpose open source solver, which gives researchers the full capability to have control not only out of the box but also inside of the “box". So APFoam can be a useful tool of broad interest in atmosphere science.
However, the merits of APreactingFoam and APSteadyReactingFoam modules (two important modules of the APFoam-1.0) are not well demonstrated/articulated. Fully coupling (two way coupling) is adopted in both APreactingFoam and APSteadyReactingFoam, where the reaction heat is considered to have impact on fluid flow. Intuitively speaking, the concentration of pollutants is too low to have a significant impact on the fluid field. Whether the heat source from reaction is considered or not might NOT have any significant effect in terms of simulation accuracy: whether to consider or ignore such tiny effects won’t change the simulation results too much. But it will have a significant effect on computational cost affecting the speed of simulations. Two way fully coupled model requires solving coupled governing equations with more unknown variables, which usually requires much more computational resources. In addition, the fully coupled system has more constraints on time steps and hence needs smaller time steps. The numerical algorithms become more complicated, too. As a general tool (calculation framework) for doing simulation, computational efficiency is important. Complicated model also makes results analysis more

complicated as there are more factors that need to be considered. In that sense, the simplest 3D module, APonlyChemReactingFoam, might be the best choice for most situations due to the following reasons:

- Faster in term of simulation and reduce computational cost significantly as explained above.
- Flow field data can be calculated offline and reused if the CFD set up does not change. For example, in certain case study, you keep the geometry, boundary condition and initial condition unchanged and only vary the source of pollutant (such as the locations, the pollutant release rate et al), then you only need to do the CFD simulation for once and reuse it for different case studies, this will significantly speed up your case study.

Response:

We are very thankful to reviewer for his/her constructive criticisms and valuable comments, which were of great help in improving the quality of the manuscript.

1. The APFoam is based on the OpenFOAM. In the original chemistry solver of OpenFOAM, the reaction heat source is considered in the solver. Actually, except the photochemical reaction, APFoam can also calculate other reaction problems which has greater reaction heat source. Therefore, the as one of the main characteristics of chemical reactions, we still keep this feature in the model.

2. The time step for this coupled system mainly follows the CFL condition from the previous studies (Bright et al., 2013; Garmory et al., 2009; Kim et al., 2012; Kwak et al., 2013; Sanchez et al., 2016; Zhong et al., 2017). The chemistry is solved by the ordinary deferential equation (ODE) solvers in OpenFOAM library, in which the chemical reactions can be integrated by dividing the flow time step into serval sub-time steps, automatically.

3. The simulations with chemical mechanism (using APreactingFoam or APSteadyReactingFoam) are initialized by the convergent flow fields (e.g., velocity and temperature, turbulence) simulated by the original flow solver in OpenFOAM. The flow fields keep almost unchanged during the simulation with chemistry. Furthermore, the simulation cost for flow fields equations (only 6 equations) is pretty small compared to the cost for the pollutant equations (about 52 equations in CS07A). Actually, only 11% of cpu time is consumed for the flow fields during each time step. Therefore, the present the fully coupled system does not affect the simulation efficiency of the solvers.

General comment 2:

Validation of the calculation framework is validated only using APonlyChemReactingFoam. APreactingFoam and APSteadyReactingFoam are not validated in the validation section. Not clear if APreactingFoam and APSteadyReactingFoam have ever been used in the case study section or not, the author did not explicitly mention that. Do these two modules (with much more complicated

governing equations) really have certain advantages in any situation? If so, I would encourage the author to justify it in a proper way, either based on literature review or ideally with a real case study. The authors need to show that considering and not considering the heat effect from reaction will have significant effect on simulation results for certain cases. With the above being said, It would also be necessary to compare the simulation speed of those three modules. In addition, in the validation section (section 3), the only validated 3D module APonlyChemReactingFoam is not validated in a fully coupled manner. The reaction model is validated alone (section 3.1). The CFD model is validated alone (section 3.2), too. And the pollutant species transportation and dispersion validation (section 3.3) is, at least, decoupled from reaction. The model is fully coupled, but the validation is done in a decoupled manner. Maybe the coupling between reaction and pollutants is also very weak and can be decoupled as well?

In summary, the models seem more complicated than necessary and lack sufficient validation. I would recommend accepting this manuscript to publish on GMD only after the major concern and specific comments (see next section) being properly addressed.

Response:

1. We had rechecked the 2D validation case and noted that the simulation case uses APreactingFoam. We apologize for the writing mistake in the manuscript. Generally, it should use the fully coupled solver (turbulence, pollutant dispersion and chemical reactions) to obtain the accurate results when considering the case with chemical reaction. Thus, in the original chemistry solver of OpenFOAM, the turbulence equation, pollutant transport equation and energy equation are coupled in the solver due to this reason. APonlyChemReactingFoam and APSteadyReactingFoam are the solvers which are developed based on the characteristics of atmospheric photochemical reactions (the chemical reactions are almost not affect the air flow) to save the simulation time (especially APonlyChemReactingFoam).
2. In all solvers of the APFoam, the reaction heat effect is considered. As mentioned above, APFoam is based on the OpenFOAM. In the original chemistry solver of OpenFOAM, the reaction heat source is considered in the solver. Actually, except the photochemical reaction, APFoam can also calculate other reaction problems which has greater reaction heat source. Therefore, the as one of the main characteristics of chemical reactions, we still keep this feature in the model.
3. We also compared the elapsed time between three solvers and found that the total elapsed time of APonlyChemReactingFoam is the longest. However, if the flow field has been determined and no need to recalculate, APonlyChemReactingFoam can save 11% of elapsed time compared with APreactingFoam while running the same setting case.
4. Due to the rarely wind tunnel experiments with chemical reactions, the model is only validated separately. We will continue to follow up the research on model accuracy in the future.

2 Specific comments

Specific comment 1：

Abstract, line 9. Numerical "resolution", in the context of mesh-based methods, such as the finite volume method used in the paper, depends on grid size, which is purely a choice in preprocess. Not clear to me how the framework developed in this paper can "improve the resolution". By "resolution", do you mean to say that the framework you developed targets at modeling small scale phenomena (such as street scale flow)?

Response:

Thanks a lot for pointing out this. The “improve the resolution” here means that the model can obtain the flow and pollutant dispersion in smaller scale. To make the expression clearer, we revised the sentence in the manuscript line 8-11:

Urban air quality issue is closely related to the human health and economic development. In order to investigate the street-scale flow and air quality, this study developed the Atmospheric Photolysis calculation framework (APFoam-1.0), an open-source CFD code based on OpenFOAM, which can be used to examine the micro-scale reactive pollutant formation and dispersion in the urban area.

Specific comment 2：

Abstract, line 14. The framework is also validated against SAPRC box modeling software, which is an essential validation, why only mention the wind tunnel validation in the abstract? Worthwhile to mention both validations.

Response:

Thanks a lot for the useful hint. The revision was done in the Abstract line 13-15:

Additionally, the model including photochemical mechanism (CS07A), air flow and pollutant dispersion has been validated and shows the good agreement with SAPRC modeling and wind tunnel experimental data, indicating that the APFoam has sufficient ability to study urban turbulence and pollutant dispersion characteristics.

Specific comment 3：

Abstract, general comments. Please double check the grammar in the abstract.

Response:

Thanks a lot for your mention. We had polished the grammar in the abstract and some grammatical

mistakes are corrected in the abstract.

Specific comment 4：

Section 1, general comments. It would be helpful for readers to have a better understanding on the major contribution of this paper if you have a little bit more detailed discussion regarding existing CFD based simulation studies in this area. For example, do they consider atmospheric photochemical in their simulation? Are those five reactions recently added in this paper already been studied in other research? In the existing studies, do they consider two way coupling or one way coupling? What tool do they use in their simulation study, OpnFoam or commercial softwares?

Response:

Thanks a lot for pointing out this. We have revied the introduction and added the information including photochemical mechanism, research parameter and CFD platform about the previous studies in Table 1. Many studies have not clearly pointed out whether their coupling method is one-way or two-way, so we have not sorted it out yet. The revision was done in the manuscript line 67-81:

With the rapid growth of the high-performance computing (HPC) platforms, computational power is no longer an obstacle. CFD simulation shows the good application prospect in urban microclimate research (Fernandez et al., 2020; Garcia-Gasulla et al., 2020). Many CFD models coupled with photochemical reaction mechanism have been developed to investigate the street-scale air quality problem in recent years (See Table 1). More commonly, simple photochemical mechanism with only three reactions (Leighton, 1961) is adapted in CFD models. This mechanism can simulate the NOx-O3 dispersion with a lower computational requirement. Many previous studies have investigated the pivotal factors that affect the reactive pollutant distribution within the street canyon by using CFD model with simple photochemical mechanism, such as street-building aspect ratio (He et al., 2017; Zhang et al., 2020; Zhong et al., 2015), ambient wind conditions (Baker et al., 2004; Merah and Noureddine, 2019), thermal effects (Baik et al., 2007) or emissions from vehicle (Liu et al., 2018a; Zhang et al., 2019b). However, due to the simple photochemical mechanism ignoring the effect of other nitrogen oxides and VOCs on the photochemistry, some studies recently have applied the full photochemical mechanism in CFD models to reduce the uncertainty of pollutant simulation. Photochemical mechanisms contain NO_x-O_3-VOCs reactions and photochemistry, such as CBM-IV (Garmory et al., 2009; Kwak et al., 2013; Kwak and Baik, 2012, 2014), GEOS-Chem (Kim et al., 2012; Park et al., 2016), RCS (Bright et al., 2013; Zhong et al., 2017), and CCM (Sanchez et al., 2016) are successfully coupled with CFD models and applied to analyse the street-scale pollutant dispersion.

Table 1. Overview of the CFD studies with photochemical mechanism

study	photochemical mechanism	Parameter	Platform
Baker et al., 2004	simple	wind conditions	RAMS
Baik et al., 2007	simple	thermal effects	Own code
Zhong et al., 2015	simple	aspect ratio	OpenFOAM
He et al., 2017	simple	aspect ratio	Fluent
Liu et al., 2018a	simple	emissions	Own code
Merah and Noureddine, 2019	simple	wind conditions	Ansys-CFX
Zhang et al., 2019b	simple	emissions	Fluent
Zhang et al., 2020	simple	aspect ratio	Fluent
Garmory et al. 2009	CBM-IV	chemical mechanism	Fluent
Kim et al., 2012	GEOS-Chem	emissions	Own code
Kwak and Baik, 2012	CBM-IV	emissions	Own code
bright et al., 2013	RCS	chemical mechanism	RAMS
Kwak et al., 2013	CBM-IV	wind conditions	Own code
Kwak and Baik, 2014	CBM-IV	thermal effects	Own code
Park et al., 2016	GEOS-Chem	thermal effects	Own code
Sanchez et al., 2016	CCM	chemical mechanism	STAR-CCM+
Zhong et al., 2017	RCS	chemical mechanism	OpenFOAM

Specific comment 5:

Section 2, question. How do you determine the time step for this coupled system?

Response:

Thanks a lot for the useful hint. Generally, the time step for this coupled system follows the CFL condition. For reference, the time step of the simulation is between ~10^{-3} to ~10^{0} second from the previous studies (Bright et al., 2013; Garmory et al., 2009; Kim et al., 2012; Kwak et al., 2013; Sanchez et al., 2016; Zhong et al., 2017). The revision was done in the manuscript line 199-202:

Even so, the time step (Δt) generally follows the Courant–Friedrichs–Lewy (CFL) condition to maintain numerical stability, which is:

$$Co = \frac{U\Delta t}{\Delta x} \leq 1 \qquad (13)$$

where Δx is the grid size.

Specific comment 6:

Section 2.1, line 113. Missing“ boundary condition", which is included in Fig. 1 but not mentioned in this paragraph.

Response:

Thanks a lot for your mention. The revision was done in the manuscript line 119-120:

For the simulation running (see Figure 1), mesh files, configure files, initial and boundary condition files should be prepared before the simulation.

Specific comment 7:

T in Eq.1 is supposed to be the temperature of the mixture? In Kelvin or Celsius?

Response:

Thanks a lot for pointing out this. *T* is the temperature of mixture in Kelvin. The revision was done in the manuscript line 135:

where *A*, *B* and *E* are the parameters of the reaction rates, and *T* is the temperature of mixture in Kelvin.

Specific comment 8:

In Eq. 2, Question. How are the lower and upper limit of the integration determined?

Response:

Thanks a lot for your attention. The lower and upper limit of the integration (λ_1 and λ_2) in Eq. 2 are the photolysis wavelength ranges according to the specific species. The revision was done in the manuscript line 139-141:

where k_{phot} is the first order rate for the photolysis reaction; λ_1 and λ_2 are the photolysis wavelength ranges according to the specific species; $J(\lambda)$, $abs(\lambda)$ and $QY(\lambda)$ are the intensity of the light source, absorption cross section and the quantum yield for the reaction at wavelength λ, respectively.

Specific comment 9:

Eq. 1 - Eq. 5 use *k* to represent “reaction rates", while in Eq. 6, *w* is used as the reaction rate. Are those two reaction rates the same thing? If so, why using different symbols?

Response:

Thanks a lot for pointing out this. The *w* in Eq.6 is replaced by *k* to represent “reaction rates”. The revision was done in the manuscript line 172:

$$\frac{\partial \rho Y_i}{\partial t} = k_i(Y_i, T) \qquad (6)$$

Specific comment 10：

In Eq. 8, should the average of molecular weight also be an unknown variable that needs to be calculated based on the mass fraction of each species? Imaging that chemical reaction changes the mass fraction of different species and then leads to changes of the average molecular weight. Any justification why the average molecular weight is assumed to be constant. Actually the assumption that average molecular weight is constant sounds reasonable to me, as the average molecular weight change caused by reaction might be ignorable. Then the following up question is, how significant is the reaction heat source? Seems can be ignorable as well. Authors may need to prove whether they are significant or not.

Response:

Thanks a lot for your mention. The average of molecular weight is a variable that can be calculated based on the mass fraction of each specie in each time step. As the reviewer said, the average molecular weight change of the photochemical reaction is too small and can be assumed as a constant. It should be noted that, this is not a constant variable because the mass fraction of each specie is changing during the simulation.

In all solvers of the APFoam, the reaction heat effect is considered. As mentioned above, APFoam is based on the OpenFOAM. In the original chemistry solver of OpenFOAM, the reaction heat source is considered in the solver. Actually, except the photochemical reaction, APFoam can also calculate other reaction problems which has greater reaction heat source. Therefore, the as one of the main characteristics of chemical reactions, we still keep this feature in the model. The revision was done in the manuscript line 176-181:

where Y_i is the species mass fraction; k_i is the reaction rate; T is the temperature; h is the specific enthalpy; u_0 is the initial energy; p is the pressure; ρ is the density; $\dot{q}$ is the heat from reaction; R is the gas constant and M_{ave} is the average molar weight which can be calculated based on the mass fraction of each species during the simulation.

Specific comment 11：

Section 2.3. For the governing equations (Eq. 6 - Eq. 8) of APChemForm, there are in total n+2 governing equations, I suppose the primitive unknowns in the governing equation are: *T, ρ, p, h*, Yi, there are in total n + 4 unknowns. number of equations < number of unknowns, Seems some other equations missing or not mentioned? Mathematically, the system is not closed. Or the author assumes that two of those *T, ρ, p, h* are not unknown? Which two?

Response:

Thanks a lot for pointing out this. The governing equation has been added in the manuscript. Besides, in the model, one of the ρ and p can be set as a constant according to the needs of research (the pressure was set as constant during the present study). The other can be calculated by ideal gas

equation of state.

The expression of the governing equations is rewritten to make it clearer and the revision was done in the manuscript line 172-181:

$$\frac{\partial \rho Y_i}{\partial t} = k_i(Y_i, T) \tag{6}$$

$$h = u_0 + \frac{p}{\rho} + \int_0^t \frac{\dot{q}}{\rho} d\tau \tag{7}$$

$$h = \sum Y_i \left(\Delta h_{f,i}^0 + \int_{T_0}^{T} C_{p,i}(T') dT' \right) \tag{8}$$

$$p = \frac{\rho RT}{M_{ave}} = \sum p_i = \sum \frac{Y_i}{M_i} \rho RT \tag{9}$$

where Y_i is the species mass fraction; k_i is the reaction rate; T is the temperature of the mixture; h is the specific enthalpy; u_0 is the initial energy; p is the pressure; ρ is the density of the mixture; $\dot{q}$ is the heat from reaction; $\Delta h_{f,i}^0$ and $C_{p,i}$ are the enthalpy of formation at reference temperature T_0 and the constant-pressure specific heat (a function of temperature) of species *i*; R is the gas constant and M_{ave} is the average molar weight; p_i and M_i are partial pressure and the molar mass of species *i*. Besides, one of the p and ρ should be set as a constant for simulation according to the needs of research. The other is calculated by Eq. 9.

Specific comment 12:

Section 2, general comments.

- Symbols that are used in this paper only need to be specified for once, for example, ρ is density, you only need to explain it the first time it appears
- Need to make sure that the same symbol has consistent meaning across the manuscript, make sure the same variable is only represented by one symbol.
- There are several dummy assumptions made in the model: such as, the mixture (mixture of air and pollutants, air itself is mixture) is in a thermal dynamic and dynamic quasi static state, they all share the same temperature and velocity. Maybe worthwhile to explicitly state the assumptions that you made when you establish the model.
- Would be better to explicitly specify that, ρ is density of the mixture. Similar for *U* and *T*.

Response:

Thanks a lot for the useful hint.

1. We had checked the symbols and removed the repeated description in the manuscript.

2. We had double checked and make sure that the same symbol has consistent meaning across the manuscript. The *w* in Eq.6 is replaced by *k* to represent "reaction rates".

3. Currently, the assumption that the mixture share the same temperature and velocity is a commonly used method in CFD model for reacting flows (Haworth, 2010).

4. The revision has done in the manuscript. All the variables have added detailed and necessary descriptions.

Specific comment 13：

Eq. 10 and Eq. 20, the physical viscosity is also considered. For air, physical viscosity is much smaller than turbulent viscosity and usually ignored in air flow simulation.

Response:

Thanks a lot for your mention. We choose to keep the physical viscosity term to ensure that the equation is complete.

Specific comment 14：

Eq. 20, k shows up again here, what does k represent in this equation, recall that k was used as reaction rate in section 2. This is really confusing.

Response:

Thanks a lot for pointing out this. the k in Eq. 20 is the turbulent kinetic energy in k-ε model. As mentioned in the question below, Eq.18 – Eq.21 are essentially governing equations Eq. 13 - Eq. 15. We had removed the Eq.18 – Eq.21 in order to reduce repetition and make the manuscript clearer.

Specific comment 15：

Section 3.2. May worthwhile to explicitly state that the Eq. 18 - Eq. 21 are not a new set of governing equations, they are essentially governing equations Eq. 13 - Eq. 15 with turbulence model.

Response:

Thanks a lot for your mention. We had removed the Eq.18 – Eq.21 in order to reduce repetition and make the manuscript clearer.

Specific comment 16：

Section 3.2, line 256. Do you treat the air as incompressible flow in the simulation?

Response:

Thanks a lot for your attention. The air flow is treated as incompressible flow in the simulation. The revision was done in the manuscript line 296:

The air flow is assumed as incompressible steady-state turbulent flow in the simulation.

Specific comment 17：

Section 3. In the validation case, what is the Reynolds number for the CFD simulation set up? It is not clear to me why you choose to scale the geometry in CFD simulation? Why not use the same geometry as whatever in the experiment?

Response:

Thanks a lot for the useful hint. The *Re* number in full-scale flow CFD validation (*H/W* = 2.4, *H* = 24m) is about 2.14×10^7, and that in wind-tunnel-scale experiments (*H/W* = 2.4, *H* = 0.12 m) is 1.9×10^5.From the previous study, the critical of the Reynolds-number-independence is about 8.7×10^4 with the aspect ratio (*H/W*) of 2 (Chew et al., 2018; Yang et al., 2021). Thus, the *Re* number in both wind-tunnel-scale and full-scale models satisfy the requirement of Reynolds number independence. The normalized wind profiles with two scales can be compared for the validation purpose. Such validation technique has been adopted in the literature (Hang et al., 2020; Yang et al., 2021).The revision was done in the manuscript line 284-288:

The corresponding Reynolds number ($Re = \frac{U_{ref}H}{v}$) in full-scale flow CFD validation (*H/W* = 2.4, *H* = 24m) is about 2.14×10^7, and that in wind-tunnel-scale experiments (*H/W* = 2.4, *H* = 0.12 m) is 1.9×10^5, which satisfy the requirement of Reynolds number independence (the critical is about 8.7×10^4 with the *H/W* of 2) (Chew et al., 2018; Yang et al., 2021). The normalized wind profiles with two scales can be compared for the validation purpose. Such validation technique has been adopted in the literature (Hang et al., 2020; Yang et al., 2021).

Specific comment 18：

Section 3. Why no 3D simulation attempted? Any difficulties or due to computational cost?

Response:

Thanks a lot for your mention. The 3D simulation is added in section 3.4. The revision was done in the manuscript line 340-364:

3.4 Pollutant dispersion in 3D street canyon

As mentioned in section 3.3, 3D pollutant dispersion validation with tracer gas is conducted in this study, following the pervious study (Zhang et al., 2019b). Simulation results also compares with the wind tunnel experimental data (Chang and Meroney, 2001). CFD domain configuration is presented in Figure 9a. In this case, six buildings are set in the domain. Building height (H) and street canyon width (W) is 0.08 m with the *H/W* = 1. Building length (Lx) and building width (Ly) is 0.276 m and 0.184 m, respectively. The distance between buildings and domain inlet, side boundary, top boundary and domain outlet are respective 5*H*, 5*H*, 10*H* and 15*H*, for simulating a realistic results (Tominaga et al., 2008). Within the target street canyon, there are also 8 measurement points (4 of which on the leeward side and 4 on the windward side) for measuring the concentrations (Figure 9b). Besides, 6 more measurement points are also set on the top of the downstream building.

Pollutant concentrations at each measurement point in this simulation case are normalized with respect to the P5 (C_i/C_5) within the street canyon. The source of the C_2H_6 is set as an inlet at the bottom of the target street canyon. The size of the source is 0.005 m in width and 0.092 m in length setting in the middle of canyon. The release velocity is 0.01 m/s toward top boundary and the mass fraction of the C_2H_6 is 1 (pure gas of C_2H_6). For 3D pollutant dispersion simulation, APreactingFoam solver with Standard k-ε model is applied to solver compressible unsteady-state turbulent flow and pollutant dispersion as well. Photochemical mechanism is not used in the simulation. The minimum grid size in this case is 0.0005 m with expansion ratio of 1.1 from the wall surface toward surrounding. Time step of simulation is set as 1×10^{-4} s and ODE solvers for chemistry is used in this validation case as well. Meanwhile, the inlet velocity and TKE profile are also retrieved from and fitted by the experimental data (Figure 10).

Figure 11 shows the comparison results between CFD simulation and experimental data. Overall, CFD simulation in 3D dispersion case slightly overestimates the concentrations in the street canyon. As for P23 and P24, the simulated results also overestimate, effected by the higher concentrations predicted within street canyon. Similarly, Statistical variables such as NMSE, FB and R are calculated to evaluate the performance of the model. As shown in Table 3, the value of NMSE, FB and R are 0.16, -0.21 and 0.93 in the 3D dispersion case, respectively, which agrees with acceptance criteria. In general, APFoam also shows the good performance in the 3D pollutant dispersion simulation.

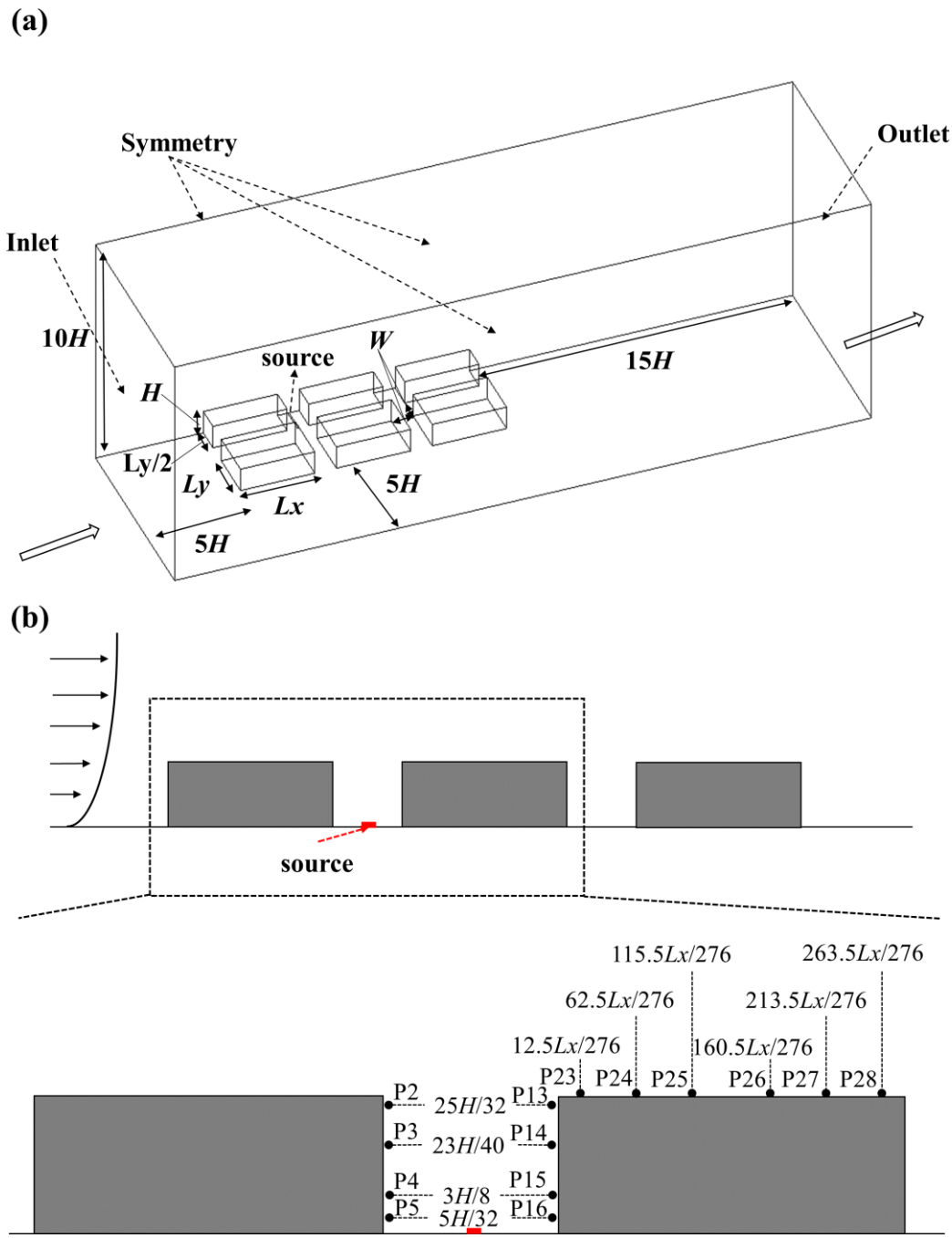


Figure 9. (a) The simulation domain of 3D pollutant dispersion and (b) the measurement points setting in the street canyon

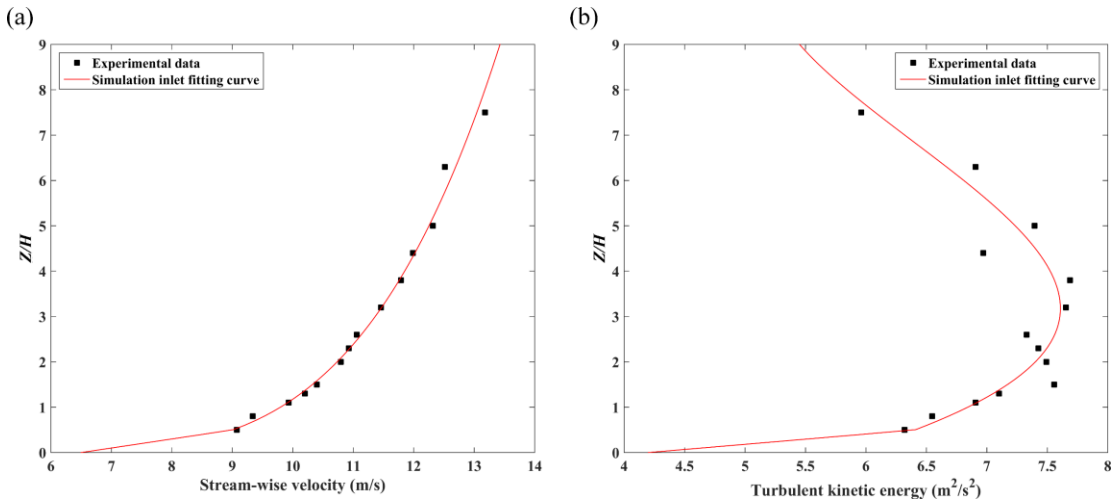


Figure 10. The inlet profile of (a) stream-wise velocity and (b) turbulent kinetic energy in 3D dispersion case

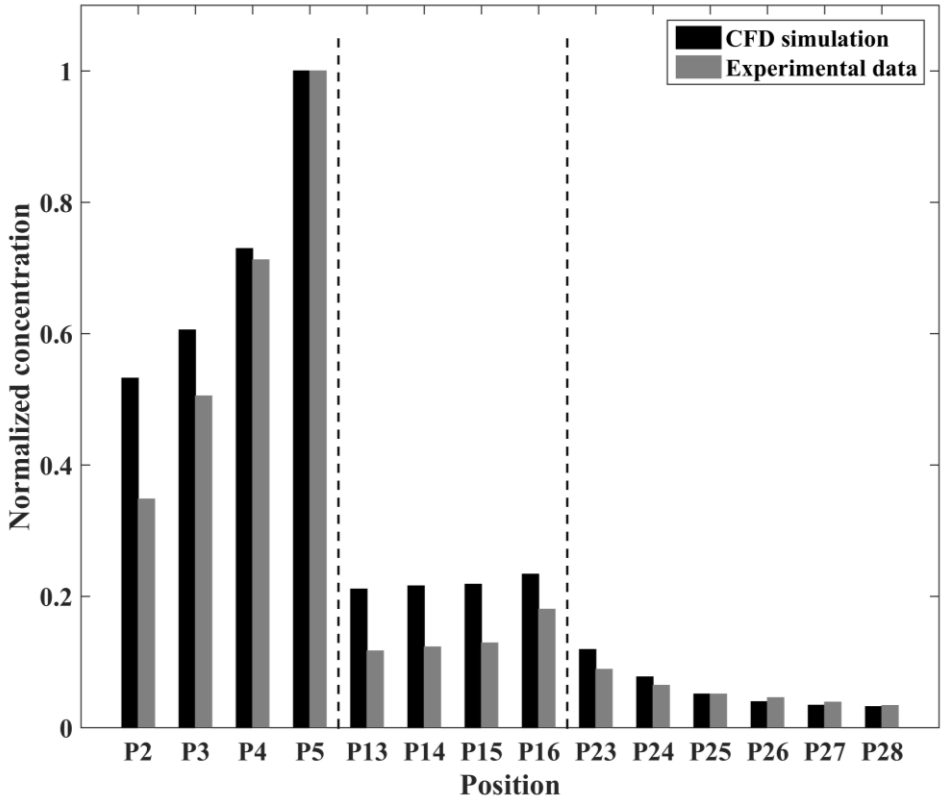


Figure 11. Normalized concentrations of CFD and experimental data at each measurement point in 3D dispersion case

Specific comment 19：

Section 3. The validation mainly validates the pollutant transportation and dispersion in the fluids field coupled with reaction, no reaction-flow-transportation coupling situation be tested. Namely, APSteadyReactingFoam, APreactingFoam are not tested. So do the authors also believe that the

effect of reaction to flow is too weak that no need to be overly concerned about it?

Response:

Thanks a lot for the hint. We had rechecked the 2D validation case and noted that the simulation case uses APreactingFoam. We apologize for the writing mistake in the manuscript.

Also the comparison among three solvers are carried and can be refer in section 4.2. The result of the comparison can refer to the response to the follow-up question.

The revision was done in the manuscript line 327-330:

For CFD simulation, APreactingFoam solver with the standard k-ε model is applied to solve the compressible unsteady-state turbulent flow field and pollutant dispersion. In order to be consistent with the wind tunnel experiments setting, photochemical mechanism is not used in the simulation.

Specific comment 20：

Section 3.3, general question. Could you clarify which of the following strategies is used in your validation?

- Solve the turbulent fluids governing equation (Eq. 18 - Eq. 21), pollutant species transportation equation (Eq. 12) and reactions (Eq.1 - Eq. 5) simultaneously.
- First do the fluids field simulation without considering pollutant species and r action, that is, first solve Eq. 18 - Eq. 21. Then do species transportation and dispersion together with reactions based on fluids field solution obtained in the first step? Namely, solve Eq. 12 and Eq. 1 - Eq. 5 in second step.

Response:

Thanks a lot for your attention. In the manuscript, turbulent fluids, chemical reaction and pollutant dispersion are solved simultaneously when the simulation cases use APreactingFoam. In the cases with APonlyChemReactingFoam, the solution of turbulent fluids governing equation is switch off, only chemical reaction and pollutant dispersion are solved during simulation. Summarily, the solution strategy is that all the equations are solve in the same time step. The revision was done in the manuscript line 186-187 and line 209-210:

For APreactingFoam, flow field, chemical reaction and pollutant dispersion are solved simultaneously in the same time step in this solver.

APonlyChemReactingFoam is only capable of solving the chemical reaction and species dispersion in the same time step under a certain flow field. The solution of turbulent fluids governing equation is switch off.

Specific comment 21：

Section 3.3, general comments. What is the time scale for reactions and what is the time scale for species transportation and dispersion? The motivation to ask this question is to see if there is possibility to decouple reaction from species transportation in such micro-scale simulation. In this section, both the experiment and the numerical simulation do not consider chemical reaction. Does this indicate that the coupling between reaction and species transportation and dispersion is ignorable as well? That is to say E_i in Eq. 12 can be removed.

Response:

Thanks a lot for the useful hint. Actually, the chemical reactions and species transportation are solved decoupled during the solvers. During each flow time step (time step for transportation), the chemistry is solved by the ordinary deferential equation (ODE) solvers in OpenFOAM library, in which the chemical reactions can be integrated by dividing the flow time step into serval sub-time steps, automatically. Then, the mean reaction rates during the flow time step will be calculated and be used in the solving of transport equations. Currently, there are rarely wind tunnel experiments with chemical reactions. Thus, the 2D and 3D validation in this study are only carried with tracer gas. We are also curious about the couple and decouple problem about the species reaction and transportation. But in generally, both chemical reaction and transportation affect the pollutants distribution.

E_i is the pollutant emitted from the source, it cannot be removed from the equation because there are emission source in validation cases. The revision was done in the manuscript line 331-333 and line 317-319:

Time step of simulation is set as 1×10^{-4} s in this validation case. The chemistry is solved by the ordinary deferential equation (ODE) solvers in OpenFOAM library, in which the chemical reactions can be integrated by dividing the flow time step into serval sub-time steps, automaticall

Currently, there are rarely wind tunnel experiments with chemical reactions. Thus, the pollutant dispersion accuracy in 2D street canyon is validated by wind tunnel experimental data with tracer gas (Meroney et al., 1996), following the pervious study (He et al., 2017b; Zhang et al., 2020).

Specific comment 22：

Section 3, general comments. Comparison between those three modules: APSteadyReactingFoam, APreactingFoam and APonlyChemReactingFoam would be very helpful.

Response:

Thanks a lot for your mention. Comparison between APSteadyReactingFoam, APreactingFoam and APonlyChemReactingFoam has been carried in section 4.2 for the $H/W = 1$ street canyon case. The

result of the comparison can refer to the response to the next question.

Specific comment 23：

Section 4, general comments/questions. Could you explicitly specify which one of the three (APonlyChemReactingFoam, APSteadyReactingFoam and APreactingFoam) is used in case studies. Any discussions to compare APonlyChemReactingFoam with APSteadyReactingFoam/APreactingFoam.

Response:

Thanks a lot for your mention. Comparison between APSteadyReactingFoam, APreactingFoam and APonlyChemReactingFoam has been carried in section 4.2 for the $H/W = 1$ street canyon case. We have compared the flow field and the pollutant distribution for these three solver results. The comparison has shown that due to the different flow algorithm, the pollutant dispersion has slightly different in the street canyon. We also compared the elapsed time between three solvers and found that the total elapsed time of APonlyChemReactingFoam is the longest. However, if the flow field has been determined and no need to recalculate, APonlyChemReactingFoam can save 11% of elapsed time compared with APreactingFoam while running the same setting case. The revision was done in the manuscript line 417-441:

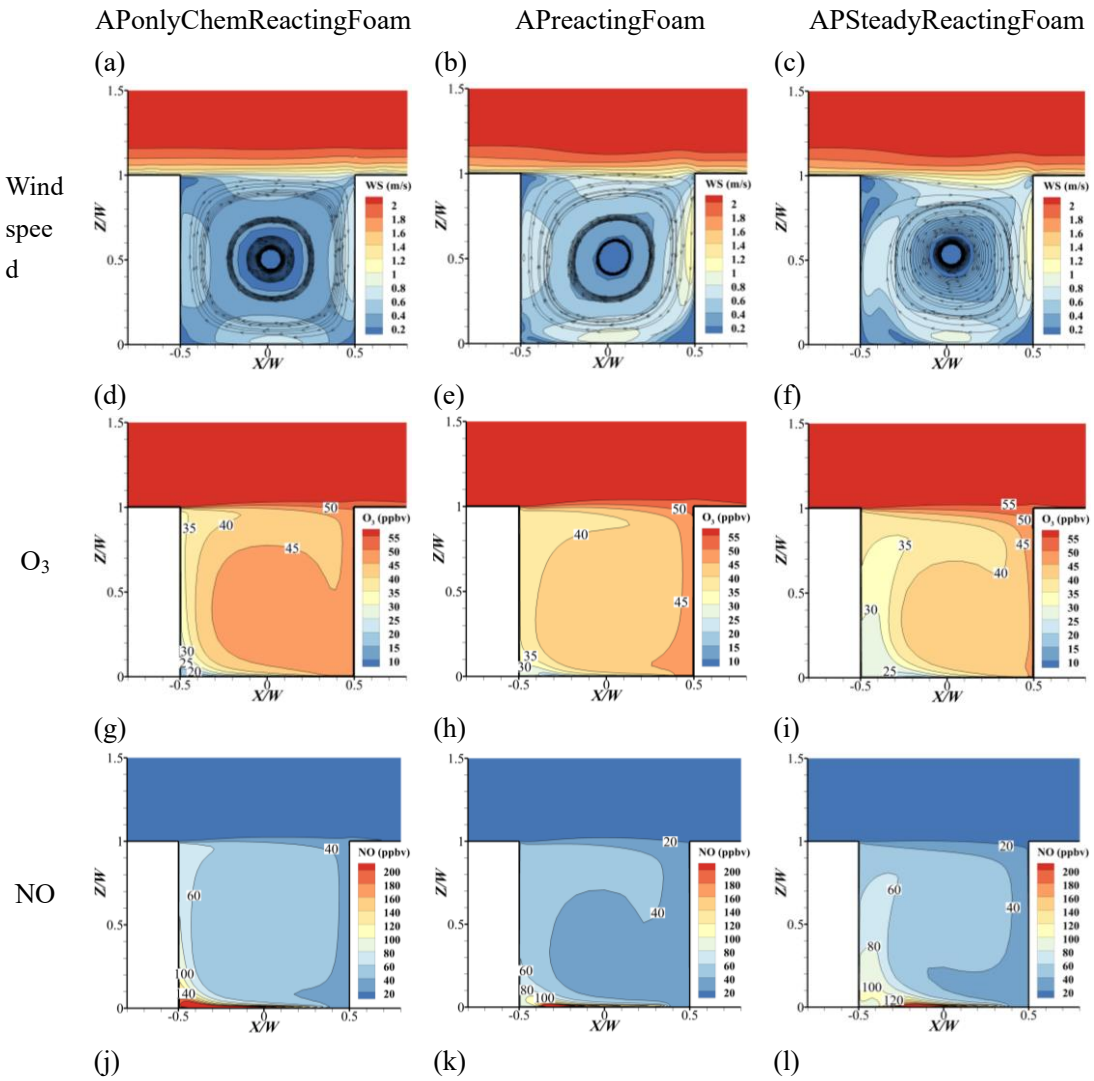
4.2 The comparison of pollutant distribution among the 3D CFD solvers

To investigate the difference of APonlyChemReactingFoam, APreactingFoam and APSteadyReactingFoam results, the comparisons of O_3, NO, NO_2 and CO distribution are conducted in $H/W = 1$ street canyon in this study. For APonlyChemReactingFoam, the flow field is treated as the incompressible steady-state flow and pre-solved using the SIMPLE method. The under-relaxation factors and residuals threshold for convergence are same as the setting in section 3.2. Chemical reaction and pollutant dispersion are solved under the steady-state flow for 90 minutes. For APreactingFoam and APSteadyReactingFoam case, turbulence flow, chemical reaction and pollutant dispersion are solved simultaneously for 90 minutes. The results in Figure 13 and all subsequent Figure are the pollutant dispersion at 90 minutes.

As depicted in Figure 13, the wind speed in APonlyChemReactingFoam case (Figure 13a) is lower than that in APreactingFoam (Figure 13b) and APSteadyReactingFoam (Figure 13c) case. The reason for the difference is most likely due to the different turbulence flow algorithm, where the turbulence is treated as incompressible steady flow, compressible unsteady flow and compressible steady flow in APonlyChemReactingFoam, APreactingFoam and APSteadyReactingFoam, respectively. Because of the slightly difference in wind speed, the concentrations of APonlyChemReactingFoam (Figure 13d, g, j, m) for pollutants are higher (due to the lower wind speed) than that in APreactingFoam (Figure 13e, h, k, n) and APSteadyReactingFoam (Figure 13f, i, l, o) case.

Table 5 shows the elapsed time of these three simulations in same case setting. Totally, the elapsed time of APonlyChemReactingFoam case (226 minutes) is slightly longer than that of APreactingFoam (214 minutes) and APSteadyReactingFoam (217 minutes) case while employing 192 CPU cores (16 × Intel® Xeon® E5-2692) for simulation. However, if the flow filed has been determined and no need to recalculate in the simulation case, the APonlyChemReactingFoam only takes 191 minutes for solving the chemical reaction and pollutant dispersion, which is 11% less time than APreactingFoam.

Many previous studies have treated the urban air turbulence as incompressible steady-state flow and investigate the pollutant dispersion successfully(He et al., 2017b; Ng and Chau, 2014; Zhang et al., 2019a, 2020, 2019b). With less time spending, the APonlyChemReactingFoam is applied in the study to analyse the photochemical reaction process in the street canyon.



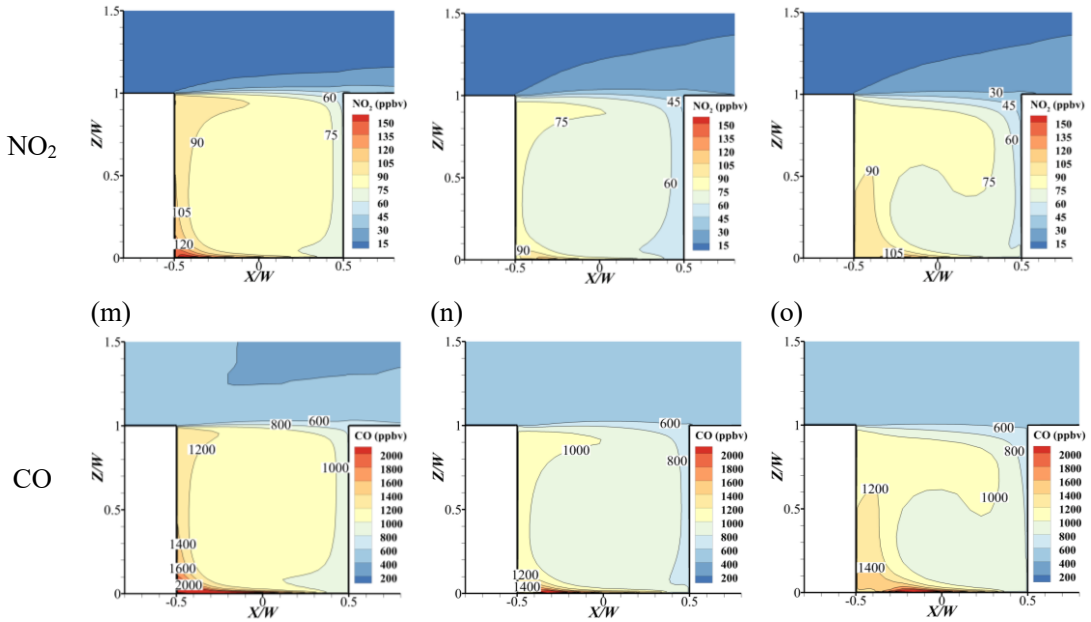


Figure 13. The comparison of (a-c) wind speed, (d-f) O_3, (g-i) NO, (j-l) NO_2 and (m-o) CO between APonlyChemReactingFoam, APreactingFoam and APSteadyReactingFoam

Table 5. The elapsed time of three solvers

	APonlyChemReactingFoam	APreactingFoam	APSteadyReactingFoam
elapsed time (minute)	191 + 35 (for turbulence)	214	217

Specific comment 24:

General comments regarding the associated source code. “README" has detailed instructions for compilation, but does not specify any dependencies, such as third party libraries that needed? Any requirement in terms of the version of these dependencies? In addition, I would recommend adding detailed instructions about how to run APFoam, either in “README" or in a separate user manual. Imagining I am a fresh user of APFoam, I would need to know which executable to execute. Before executing that executable, what kind of preparation work is needed. For example, no need to instruct users how to generate mesh, but would be necessary to mention that users would need to get the meshed geometry ready before using APFoam. A little bit more Instructions about how to make use the “APFoam_tutorials" would also be very helpful.

Response:

Thanks a lot for pointing out this. Some details has been addad in README (on Github). Since the APFoam is developed based on the openFOAM, the install recommend is similar with the original openFOAM. The necessary libraries (minimum versions) mainly include: gcc (4.8.5); cmake (3.8); boost (1.48); zlib (1.2.7); openmpi (2.1.6) or mpich (3.2.1). Additionally, a tutorial of APFoam is add in Github as well.

3 Technical corrections

Abstract, line 9. Change “newly" to “new".

Abstract, line 13. Change “reaction" to “reactions".

Response:

Thanks a lot for pointing out these problems. The revision was done in the manuscript.

In Eq. 1, please specify the meaning of T?

Response:

Thanks for your attention. T is the temperature of mixture. The revision was done in the manuscript:

where A, B and E are the parameters of the reaction rates, and T is the temperature of mixture in Kelvin.

In Eq. 7, what is the h? Specific enthalpy?

Response:

Thanks a lot for pointing out this. h is the specific enthalpy. The revision was done in the manuscript:

where Y_i is the species mass fraction; k_i is the reaction rate; T is the temperature; h is the specific enthalpy; u_0 is the initial energy; p is the pressure; ρ is the density; $\dot{q}$ is the heat from reaction; R is the gas constant and M_{ave} is the average molar weight.

Fig. 9, caption. “The probe points locations.“

Line 307. “... in the targeted street canyon.”

Line 334. Change “obtain" to “obtained".

Line 414. “... is slightly greater than that of ..."

Line 418. Again “... the street canyon .."

Line 419. "This is because that the background "

Response:

Thanks a lot for pointing out these problems. The revision was done in the manuscript.

References:

Bright, V. B., Bloss, W. J. and Cai, X.: Urban street canyons: Coupling dynamics, chemistry and within-canyon chemical processing of emissions, Atmos. Environ., 68, 127–142, doi:10.1016/j.atmosenv.2012.10.056, 2013.

Chew, L. W., Aliabadi, A. A. and Norford, L. K.: Flows across high aspect ratio street canyons: Reynolds number independence revisited, Environ. Fluid Mech., 18(5), 1275–1291, doi:10.1007/s10652-018-9601-0, 2018.

Garmory, A., Kim, I. S., Britter, R. E. and Mastorakos, E.: Simulations of the dispersion of reactive pollutants in a street canyon, considering different chemical mechanisms and micromixing, Atmos. Environ., 43(31), 4670–4680, doi:10.1016/j.atmosenv.2008.07.033, 2009.

Hang, J., Chen, X., Chen, G., Chen, T., Lin, Y., Luo, Z., Zhang, X. and Wang, Q.: The influence of aspect ratios and wall heating conditions on flow and passive pollutant exposure in 2D typical street canyons, Build. Environ., 168, 106536, doi:10.1016/j.buildenv.2019.106536, 2020.

Haworth, D. C.: Progress in probability density function methods for turbulent reacting flows, Prog. Energy Combust. Sci., 36(2), 168–259, doi:10.1016/j.pecs.2009.09.003, 2010.

Kim, M. J., Park, R. J. and Kim, J. J.: Urban air quality modeling with full O3-NOx-VOC chemistry: Implications for O3and PM air quality in a street canyon, Atmos. Environ., 47(2), 330–340, doi:10.1016/j.atmosenv.2011.10.059, 2012.

Kwak, K. H., Baik, J. J. and Lee, K. Y.: Dispersion and photochemical evolution of reactive pollutants in street canyons, Atmos. Environ., 70, 98–107, doi:10.1016/j.atmosenv.2013.01.010, 2013.

Sanchez, B., Santiago, J. L., Martilli, A., Palacios, M. and Kirchner, F.: CFD modeling of reactive pollutant dispersion in simplified urban configurations with different chemical mechanisms, Atmos. Chem. Phys., 16(18), 12143–12157, doi:10.5194/acp-16-12143-2016, 2016.

Yang, H., Lam, C. K. C., Lin, Y., Chen, L., Mattsson, M., Sandberg, M., Hayati, A., Claesson, L. and Hang, J.: Numerical investigations of Re-independence and influence of wall heating on flow characteristics and ventilation in full-scale 2D street canyons, Build. Environ., 189(October 2020), 107510, doi:10.1016/j.buildenv.2020.107510, 2021.

Zhong, J., Cai, X. M. and Bloss, W. J.: Large eddy simulation of reactive pollutants in a deep urban street canyon: Coupling dynamics with O3-NOx-VOC chemistry, Environ. Pollut., 224, 171–184, doi:10.1016/j.envpol.2017.01.076, 2017.