Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-545-RC1, 2020

Atmospheric Chemistry and Physics
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EGU
Discussions

Interactive comment on "Error induced by neglecting subgrid chemical segregation due to inefficient turbulent mixing in regional chemical-transport models in urban environments" *by* Cathy W. Y. Li et al.

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Received and published: 3 August 2020

This research presents very interesting results on a complex and interdisciplinary topic that is not yet solved, how the turbulence influence chemistry over (urban) heterogeneous emissions. The paper confirms previous results to take into account and the need to parameterize the effects of inhomogeneous mixing. The originality of the work is to quantify the errors due to the segregation of species in air quality models and its dependence on the vertical and horizontal resolutions. In that respect, I think the use of direct numerical simulation is very welcomed in the field and the reader of ACP will

appreciate this sort of research. I have written my comments section by section.

1. Introduction

I found the introduction very complete and very well-written. The aim of the paper, its research strategy and the methods are very well posed and explained. A potential improvement to gain clarity is to make a clearer distinction to the segregation of chemical species due to the state of nature of turbulence and their drivers - thermal stability, canopy-atmosphere interactions, clouds - and the one due to the heterogeneity of the surface emissions. I realize that they are closely related, but in my opinion, the reader will appreciate better the complexity of the problem if this distinction is made. From the very complete and literature review that shows the capacity of synthesis and the deep understanding of the topic by the authors, I miss the following papers: Patton, E. G., et al. (2001): Decaying scalars emitted by a forest canopy - A numerical study, Boundary-Layer Meteorology, 100, pp. 91-12 and Baker J. (2004): A study of the dispersion and transport of reactive pollutants in and above street canyonsâĂŤa large eddy simulation. Atmospheric Environment 38, 6883-6892.

2. Model description

I would like to congratulate the authors of using the direct numerical simulation technique to move forward in the complex topic of the interactions between turbulence and chemistry. This numerical technique sheds new light, and it is complementary to the large-eddy simulation technique. I have in this section the following comments:

a) The authors defined the Damkohler number as a function of the most energetic turbulent time scale. However, I was wondering on the need to define a Damkohler number to quantify the effects of turbulence at the smaller spatiotemporal scales, for instance using a Kolmogorov time scale. Chemical reactions occur at the molecular diffusion time scale, and I wonder whether it is necessary to include a physical time scale closer to this one of chemistry. I think the reader would like to hear the opinion of the authors in that respect and if their DNS numerical experiments could add extra information.

b) At section 2.2.1, the authors focus on the most extreme case of segregation, i.e. an irreversible second-order reaction. I understand their choice, but I think the reader will appreciate if they could include in their chemical mechanism a backward reaction. By so doing, the chemical will become closer to an equilibrium, and it will probably reduce the effects of the segregation. In that respect, the analytical solution for three chemical species and two reactions derived by Jonker et al. (2004) (Characteristics length scales of reactive species in the convective boundary layer, Journal of Atmospheric Sciences 61, 41-55) can be very useful to study the effect without introducing additional computational costs. Note that variations on time and on space due to presence of clouds or vertical perturbances of the radiation transfer within the urban canopy can be interested cases to bring this triad of species out of equilibrium, and therefore enhance the segregation effect again.

3. Results

- Homogeneous emission. For the sake of completeness, I think it might be interesting in Figure 2 to add an extra panel but now with a typical time average used by the air quality models ($\sim$30 minutes) to (probably) show how the small-scale fluctuations of the chemical term are also filtered out due to the time-averaging.

- Section 3.1.2. (lines 12-14). I think the authors are reporting a very interesting results regarding the more prominent minima due to the higher vertical resolution. Could they please elaborate and quantified a bit more?

- Since the paper is entitled “errors”, I think an additional figure 3 that can complete the discussion is the calculation of the exchange coefficient K defined as the ratio between the turbulent flux and the mean gradient). I realize that in the middle of the CBL can be undetermined, but close to the surface and at the entrainment zone it might provide information on the combined effect of chemistry and turbulence on the parameterization of the transport of chemical species.

- Section 3.2 (line 29). It might be interesting to show how this Is = - 0.8 changes if a first-order backward reaction is included in the chemical mechanism of reaction R1.

- Figure 9. Could the authors elaborate more the reasons of lower values on k_eff/k at the entrainment zone compared to the values near the surface? Both regions are characterized by higher gradients of the chemically active species. Why are these differences at these two regions?

4. Discussion

I think it can be worth to make a discussion point that the errors are not only on the segregation and therefore on the chemical reaction rate, but also on the eddy diffusivity and therefore on the transport of chemical species.

Section 5.1. (lines 9-16) Will look-up tables depending on the processes that they hace mentioned be a solution instead of the parameterizations?

Page 17 and line 1. Will previous work on natural canopies be useful to be connected to urban canopies?

Since section 5.2 presents an outlook of possibilities for future research, I think it might important to include a short description on the effects of radiation and its perturbation (clouds, urban canopy) closely couple to the turbulent effect. An additional experiment including the backward reaction as here proposed can pave the way to future research.

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