Response to Reviewer III

General Comments:

Qu et al. have presented and nuanced two different types of budgets for regional ozone pollution: concentration budget and mass budget. They start by mathematically formulating both types of budgets from first principles, basically using the fundamental principle of mass conservation in a Eulerian framework, similar to a continuity equation where the rate of change of concentration/mass is the sum of horizontal advection, vertical exchange and local source term (chemical production, dry deposition etc.). They then present their method for calculating the two types of budgets on WRF-CMAQ gridded model output. They have chosen the Pearl River Delta region (PRD) in China as their study region.

For the concentration budget, they break down the vertical exchange term into two separate terms: vertical entrainment/detrainment of air due to temporal changes in boundary layer height (ABLex-H) and horizontal advection of air through the extra volume of air created due to increasing boundary layer height (ABLex-M). Since the study region is not a perfect cube, they have defined four boundaries roughly corresponding to north, east, west and south directions, like four sides of a cube, to deal with the transport in a Eulerian framework. To calculate the transport contribution to the change in concentration in the boundary layer, they make use of the concentrations of the horizontal advecting air mass as well as the background concentration of the air above the boundary layer in the region. Similarly, transport contribution in the mass budget is calculated by adding the new mass brought in through advection and vertical exchange.

The key point here is that often new mass is added from non-local sources through transport but this increase in mass is simultaneously accompanied by an increase in boundary layer volume which diminishes any considerable increase in concentration within the boundary layer. Therefore, these non-local contributions are diminished in the concentration budget although the composition of the pollution has changed, i.e., there are more O_3 molecules in the region from the non-local sources without any (or much) change in concentration.

The authors then perform 3 different sensitivity simulations where they zero-out emissions for the PRD region, Eastern and Central China region but not PRD, and all regions within inner model domain, respectively. Using the difference between these sensitivity simulations and the baseline run, they calculate the contributions of these source regions to the O_3 mass and O_3 concentration in the PRD region. In Figure 3 they show that the change in mass is driven in large amount by vertical entrainment but this addition of transported mass in the morning is accompanied by increase in boundary layer volume and the removal of transported mass in the evening is accompanied by a decrease in boundary volume such that the transport does not have a large effect on concentration budget. They further show in Figure 6 that a major part of vertical exchange and horizontal transport in the mass budget comes from non-local and background sources, and that the horizontal transport is greater than local chemical production in autumn and the opposite in summer.

Overall, the authors have highlighted an important point on mass contributions of O_3 (or any other longer-lived pollutant) which gets concealed in concentration budgets due to volume changes in boundary layer. Mass budget might become more important than concentration budget particularly in cases when the chemical species in consideration has a different characteristic (e.g., toxicity) based on its source region. I recommend this manuscript for publication with minor corrections.

Response:

We appreciate your positive comments on our paper. Following your summary of our contents in the general comments, for clarity, we have modified the abstract, in lines 33-38 (the line numbers used correspond to those in the revised manuscript with author's changes; same below):

Through high contributions to the O_3 mass increase in the morning, transport determines that most O_3 in the PRD originates from the global background and emissions outside the region. However, due to the simultaneous rapid increase of ABL volumes, this process only has a relatively limited effect on O_3 concentration increase compared to photochemistry, and transport effect on the regional sources of O_3 cannot be illustrated by the O_3 concentration budget.

and the revised manuscript, in lines 551-553:

However, accompanied with the simultaneous rapid increase of ABL volumes, this process has a relatively limited contribution to O_3 concentration increase in comparison to photochemistry.

and in lines 574-578:

Massive O_3, mostly derived from non-local sources, being transported into the ABL in the morning has a relatively limited influence on the O_3 concentration increase (25% and 5% in autumn and summer, respectively) compared to photochemistry because of the rapid change of ABL volumes at the same time. However, this process nearly determines the dominance of non-local source contributions for daytime O_3 in the PRD.

The manuscript has also been revised based on other suggestions. Please find below our responses to the specific comments (in blue) and corresponding revisions (in red).

Specific comments:

1) Include the domain map showing at least d02 of WRF-CMAQ with clear demarcation of the different source regions used in the BFM simulations.

Response:

In the original manuscript, the domain map was displayed as Fig. S5 in Supplement. We agreed that such information might be important for readers. Thus, it is now shown as the new Fig. 2 in the manuscript. All figure numbers have been corrected accordingly.

2) The names ABLex-H and ABLex-M aren't intuitive. I do not understand why those letters (H and M) were used as they can confuse the reader. I suggest calling them ABLex-A (advection through boundary layer change) and ABLex-E (entrainment through boundary layer change).

Response:

We accept the suggestion and try to express these processes in a more reader-friendly way:

The vertical exchange near the ABL top due to large-scale air motion is a process due to the advection perpendicular to the ABL top and its slope. We agree that "ABLex-A" is a better short term to indicate the process, thus all "ABLex-M" in the manuscript, tables and figures were revised into "ABLex-A". Besides, in the full term, "due to large-scale air motion" may not be clear for readers, thus it has been revised into "due to advection perpendicular to the ABL top and its slope" in the manuscript.

The vertical exchange near the ABL top due to the changes in ABL heights occurs only linked to the increases and decreases of ABL heights. It is a part of the process of vertical exchange near the ABL top, or entrainment and detrainment. Thus, we prefer to keep the short term of the process as "ABLex-H", where "H" in this manuscript is used as the parameter of ABL height in this paper. We added the note to indicate ABL height in the full name of the process is represented by "H" before introducing the short term, in lines 155-156:

… 1) the temporal changes of ABL heights (*H*) and 2) …

It is also needed to clarify that vertical exchange near the ABL top is the process of entrainment and detrainment. Thus, we added some necessary notes in relative parts, including lines 74-75:

…2) vertical exchange through the ABL top (entrainment and detrainment, the third term) …

and lines 154-155:

The terms on the right side of Eq. (3) suggest that vertical exchange through the ABL top, or entrainment and detrainment, is attributed to …

3) In this work, the authors have formulated their equations to calculate "change" in concentration and mass over time but there are plenty of studies which perform BFM-type sensitivity runs where they alter emissions over different regions and subtract the result from the baseline run to derive hourly concentrations (instead of hourly change in concentrations) attributed to emissions from that region. The authors should discuss the validity of such results and their implications for policymaking.

Response:

We thank the reviewer for the comment. To better clarify: It describes the application of BFM, a typical method of O_3 source apportionment, which aims to identify the contributions of emissions originating from different regions to the O_3 level in the targeted region. Indeed, there are several BFM studies (e.g., Wang et al. (2006) and Streets et al. (2007)) that are used for policymaking.

Our study does not question the validity of such O_3 source apportionment studies or O_3 concentration budget analysis, but suggests appropriate ways on how to apply these methods effectively: i) To lower the overall O_3 levels and achieve long-term air quality improvement, based on the results of O_3 source apportionment, it is needed to focus on emission reduction within larger areas for regions that are notably influenced by upwind sources; ii) To lower the peak O_3 levels of the day and achieve short-term alleviation of O_3 pollution, owing to the quick response of O_3 concentration increase to local emissions in the O_3 concentration budget, reducing local emissions is a better strategy. The choice of strategy to apply should depend on the specific goal of O_3 pollution control, which is set based on the effects of O_3 pollution on human health, the ecosystem, etc. Relative discussions can be found of in the final paragraph of this manuscript, in lines 603-614:

The present study concluded that transport and gas-phase chemistry play the main role in the O_3 mass and concentration budgets, respectively. As a consequence of our assessment, the following is suggested for policy-makers. For areas where non-local emissions notably contribute to O_3, emission reduction in the upwind regions can reduce the overall O_3 concentrations effectively, which is a crucial step towards the long-term improvement of regional air quality. However, for short-term air pollution control, this strategy is not efficient because emission reduction in upwind regions may need to start days earlier before the polluted periods. In contrast, reducing local emissions is expected to lower the rapid daytime O_3

concentration increase efficiently and, thereby, O_3 peak levels in the short term, as highlighted by the O_3 concentration budget. The choice of the better strategy to be applied should depend on the specific objectives of O_3 control (mean levels vs. peak levels; long-term vs. short-term), which are set based on a more in-depth understanding of O_3 effects on human health, crop yields and ecosystems. More efforts are required to systematically evaluate the effects of different emission reduction strategies on alleviating the detrimental effects of O_3.

Additional Statement:

During the validation of the revised manuscript, the ACP team noted that:

Please make sure that the lists of corresponding authors in the system and the manuscript file match.

The corresponding authors of this paper are Xuesong Wang and Yuanhang Zhang. However, in the submitting system, Xiao Teng was automatically assigned as the corresponding author and we cannot cancel it. Please note that this is not correct and need to be corrected.

Reference

Streets, D. G., Fu, J. S., Jang, C. J., Hao, J., He, K., Tang, X., Zhang, Y., Wang, Z., Li, Z., Zhang, Q., Wang, L., Wang, B., Yu, C., Air Quality during the 2008 Beijing Olympic Games, Atmos. Environ., 41(3), 480–492, https://doi.org/10.1016/j.atmosenv.2006.08.046, 2007.

Wang, Z., Li, J., Wang, X., Pochanart, P., Akimoto, H., Modeling of Regional High Ozone Episode Observed at Two Mountain Sites (Mt. Tai and Huang) in East China, J. Atmos. Chem., 55(3), 253–272, https://doi.org/10.1007/s10874-006-9038-6, 2006.