

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 O₃ 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 O₃ mass and O₃ 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 O₃ (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:

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.
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).

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.