Misrepresentation of Formaldehyde in the Model

Carbonyls act as radical sources important for the photochemical production of PM2.5 during wintertime inversion episodes in the Salt Lake Valley. The photolysis of these compounds, including formaldehyde, may be important for daytime generation of radicals, as shown by recent observations. However, although formaldehyde is important for PM2.5 formation, it may be underrepresented in the model during daytime hours, as shown by a comparison of modeling outputs to measurements conducted in winter 2017 at the University of Utah (2017 Utah Winter Fine Particulate Study (UWFPS)). The modeling results were compared to observations conducted in winter 2017 since measurements of VOCs species were not available during 2011. However, while these field study measurements from 2017 cannot be directly compared to day-specific 2011 model simulations, they’re qualitatively useful to assess if the model predicts similar levels of VOCs during strong inversion conditions.

On average during peak PM2.5 exceedance days, measured formaldehyde peaked at about 3 ppb around 11 am (Figure 2) while modeled formaldehyde peaked at 6 pm and displayed a concentration of only 1.8 ppb (figure 1) at 11 am. Modeled formaldehyde also exhibited a temporal trend different from that of measured formaldehyde, with observations indicating direct emission as well as secondary production of formaldehyde. Similarly, modeled acetaldehyde displayed a temporal trend different from that measured on peak PM2.5 days. This comparison suggests that acetaldehyde and formaldehyde, an important source of radicals, may be underestimated in the model during mid-day hours. Given the role of formaldehyde in the generation of radicals, an underestimation of formaldehyde in CAMx may increase the model’s sensitivity to oxidants and may limit its sensitivity to NOx emissions.

Figure 1. Hourly time series of average modeled formaldehyde and acetaldehyde during January 6-8 2011 at the University of Utah.

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To further investigate the model performance, UDAQ compared modeled Ox, defined as the sum of O3 and NO2, to that measured at the University of Utah and Hawthorne monitoring sites, where measurements were retrieved from the 2017 UWFPS. Similarly to formaldehyde, Ox is misrepresented in the model during daytime hours. At both Hawthorne and the University of Utah monitoring sites, measured Ox peaked in the early afternoon (Figure 3) while modeled Ox peaked in the late afternoon (Figure 4).
Figure 4. Average hourly time series of modeled Ox (NO2+O3) during January 6-8 2011 at the University of Utah and Hawthorne.

The model’s sensitivity to formaldehyde emissions was further evaluated by conducting a modeling sensitivity run where formaldehyde emissions from all sectors were increased by 50%. Formaldehyde emissions from the 2019 inventory were considered for this sensitivity simulation. Both modeled ozone and nitrate increased after increasing formaldehyde emissions, further suggesting the model’s increased sensitivity to oxidants, which may limit its sensitivity to a reduction in NOx emissions (Figure 5). An underestimation of formaldehyde results in an underestimation in the production of HNO3, leading to a reduced response to proposed NOx controls.
Figure 5. Spatial plots of the difference in mean ozone and nitrate levels between the sensitivity modeling run, where formaldehyde 2019 emissions were increased by 50%, and the 2019 emissions modeling run, where formaldehyde emissions were kept unchanged. Plots are shown for January 7 2011.