Interactive comment on "Photochemical impacts of haze pollution in an urban environment" by M. Hollaway et al.

Anonymous Referee #1

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This Hollaway et al. manuscript is tightly written and well put together. As stated in their Discussion (P12/L20), this new work "highlights the sensitivity of the responses in photolysis rates not only to the vertical distribution of aerosol but also the chemical speciation of the particulates"

The measurements of aerosol pollution in the north of Beijing combined with the careful modeling are able to demonstrate the accuracy of the modeling of photolysis rates and hence the ability to use a model to calculate the aerosol-pollution influence on near surface ozone. Their conclusion seem solid and important in understanding just how aerosol pollution interacts with oxidants. I recommend publication after the authors consider the minor suggestions below.

The impact of this work would be greater if some editorial aspects are cleaned up.

C1
P1/Abstract. I urge you to drop the decimal place on these per cents. IFor example, think that 40-60% adequately describes 40.4-66.2%. Also, I think that the key statement quoted above (P12/L20ff) should appear in the abstract. This is a powerful result and should be up front.

P2/L5 I would have expected the original 2 papers that calculated the aerosol photolytic effects to be noted here: Martin et al, and Bian et al., both 2003.


P4/L1 I read through the Whalley 2018 paper and looked up their supplemental data and cannot find any source of cross section data for photolysis. Is this the correct source?

P4/L13 does non-refractory aerosol include semi or partly volatile SOA?

P4/L31 Eqn I am trying to understand the units here. b (extinction) should be in 1/Mm, right? But [X] is usually a concentration unit (#/cm3). So please assign units carefully to all to help this reader.

P5/L3-12 Very nice design. I like the careful merger to get consistent measurement data for the modeling.

P5/L19 'account for' does not make sense to me, do you mean "average over"? Can you specify what options/versions you used in Cloud-J, IF you implemented observed cloud fraction? Cloud fraction is not mentioned here, so make it clear that you just used a single column atmosphere, full cloud or clear in each layer.

P5/L21-33 This is a nice augment to the Fast-J code, both NO2 absorption and the aerosol cross sections. Are they available as a mod to a Fast-J/Cloud-J version? They ought to be. Using H-G and asymmetry parameter is OK for generating the phase function, but Mie would be better (outside the scope of this paper I know).

P6/L8-27 The high-frequency comparison in Figure 1 is fascinating and it is interesting
to see the mean bias over the diurnal cycle. Since you are using the same cross sections for both, it means that FJX is too hot, and overestimates the high-sun fluxes. Can you check if this holds for clear, unpolluted days?

Also, I think it would be valuable here to add something like a correlation coefficient to test if FJX+'Observed aerosols' can match the daily variability. Just add the r² to the right-hand figure.

Use of %: Here is where the decimal point in the % numbers makes some sense (i.e., the bias). There are so many % numbers in this paper, it would be good to try to differentiate them simply. Otherwise the unit '%' should be fleshed out to say '% of what'. The % in a bias should maybe always have a sign: e.g., L13, +5.6 % mean bias above observations. See next section.

P6/L32 Use of %: Maybe do not need % here, but always need units to be clear. "In winter (NH₄)₂SO₄ (39% of all aerosols by mass? by number? by optical depth?) and BC (30%) provide…"

P7/L2 & Fig 2 "Vertical AOD profiles" and Fig 2 (bottom) make no sense in terms of units and what is plotted. The top row of Fig 2, it is the fraction of AOD from each component. Here the extra decimal point makes sense and does not clutter the reading. The problem is that AOD is always an extrinsic quantity while extinction (b, beta) is intrinsic. AOD is always integrated over a column or path length, but b is local. Thus you can plot b vs altitude, but not AOD. Please fix units in plot. See also L17 with "AOD values", and L20.

P7/L9 Use of %: here and elsewhere in this discussion, please round off. 20% is just fine instead of 19.8%, since the uncertainty is certainly greater than +-1%.

P7/L27 Use of % again: We have 23.8 and 23.1% reductions, the extra decimal is not meaningful in this discussion. Also the units are not clear. J’s are about -23 % below what? a clear sky? a sky with all the other pollution but only those removed? If the
latter, it does raise the question of linearity or interference across the aerosol mix.

All throughout this discussion the nearest % is more than adequate. P8L16 & Fig 4. I wonder if this figure should show absolute changes in J’s instead of %. I would think that this would emphasize the reactivity better, since % changes at low sun are not really important. For something like J-HONO, this would be fine, since the J’s are more of a square wave. Figures 3 & 5 are fine as is, even if you change this to absolute deltas. If you want, you could do % of some noontime mean.

P8L30-35 Yes, this is an important result. Aerosol-pollution scattering and absorption above the boundary layer could be the most important factor.

P9/L5+ You can note here that – as you have found for polluted boundary layer - clouds have a much greater relative impact on J[NO2] vs J[O1D] also in observations over the clean remote Pacific [Hall et al., acp-18-16809-2018, very new paper, also using FJX, not available at time of drafting this paper.]

P9/L11-17 I found this paragraph confusing, and could not get the message.

P9-P12 Discussion This section is long and to me it wanders. If this discussion is useful, please do some numbered sub-sectioning for the reader.

P10/L27 "would be balanced by . . . rise ion NO . . ." To me this is not logical, since more NO means that less of O3 is tied up as NO2 in the NOx emissions, and further, more NO would enhance the ROO+NO reactions? Does this not augment the enhanced production rates? as oppose to balance them?

P11/L6 Use of %: "around 12.0%", really. "by 12 % and 3 %, respectively."

P13/L10 It seems that I have read something like this before. Do you need to repeat?

P13/L27 I think you need to have both observational data, plus the FJX code (that part that was adapted to NO2) and aerosol scattering data for FJX. I hope you get a doi eventually because otherwise it would be impossible to find on the CEDA site.