

Interactive comment on
**“Preindustrial-to-present-day radiative forcing by
tropospheric ozone from improved simulations
with the GISS chemistry-climate GCM” by D. T.
Shindell, et al.**

D. T. Shindell, et al.

Received and published: 1 October 2003

We thank the reviewer for his comments, which we found very useful in revising the paper. In addition to many relatively minor comments, which we have addressed as detailed below, he suggested that the comparison of surface ozone with observations be greatly expanded and a figure added on this topic (something the other reviewer mentioned as well). We agree that this aspect of the paper was relatively weak, and have followed this suggestion, performing a comparison with the full Logan climatology and adding a figure showing some of the most interesting and representative sites in the comparison. Of particular interest is that the very low concentrations simulated by the model over South America and Africa appear to be fairly reasonable. The model

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agrees very well with observations from Venezuela and reasonably well with those from Cuiaba (though the amplitude of the fall biomass burning peak is underestimated). There is an annual average bias of about +5 ppbv at Natal, and -5 ppbv at Brazzaville, while biases are only -1.6 at Venezuela and +0.1 at Cuiaba, so it does not seem that there is any systematic bias over these continents. Of course, more data from these regions would be helpful. In any case, we are grateful to the reviewer for his suggestion to include this material, which we believe has greatly increased the value of the model validation presented here.

Specific comments:

Page 3941: We've added the suggested reference.

Page 3943: This is a good point, and we've now revised the discussion of the chemical mechanism to explain that the scheme we are using is a version of the CBM-4 which was modified for use in global models by Houweling et al (1998). This was done by removing aromatic compounds and adding in reactions important in background conditions, including organic nitrate and organic peroxide reactions, and extending the methane oxidation chemistry. The revised scheme was then readjusted based on the more extensive Regional Atmospheric Chemistry Model (RACM) (Stockwell et al., 1997), and the modified scheme includes several surrogate species designed to compensate for biases relative to the RACM mechanism. The modified scheme was then evaluated, and shown to agree well with the detailed RACM reference mechanism over a wide range of background chemical conditions including relatively pristine environments (Houweling et al., 1998).

Page 3949: The reviewer is correct about the 950 hPa data. We've added a description of this in the text.

Page 3948: The reviewer's insight was all too clear in this case. While this paper was in review, we discovered a programming error in the implementation of the stratospheric ozone seasonally varying climatology. We've fixed this flaw, and noted it in the text.

Given that the biases at this level are still only a few percent (Table 3), and the climatology doesn't change from the preindustrial to the present-day, we expect that this error will only have a minor impact on our radiative forcing calculation.

Page 3949: As discussed above, a comparison with surface observations has been added. Also, we note now that in addition to the average bias improving (which hides opposing positive and negative errors), the average difference between the model and the observations has also improved.

Page 3950: We've added data comparisons with Izana (based on the Logan climatology) for the N. Atlantic, and corrected the '2 ppbv' to '20 ppbv'.

Page 3951: The reviewer is correct about the difference between isoprene explicitly and implicitly as CO. We've rephrased this text to indicate that we meant the new presence of explicit isoprene, which can react directly with O₃, was important over the tropical forests or other low-NO_x regions.

The reviewer's comment about the chemistry budget is certainly true, and we now point out that the net is the difference between large production and destruction terms, so limited in value. We're referring to the overall ozone budget here, however.

The reviewer points out that the buffering of the system is not surprising as tropospheric ozone is roughly in steady-state, so increases in one term must be balanced by decreases in another. We agree with these comments, and so we've revised our discussion along these lines. We've refrained, however, from giving the budget numbers in a table, as we wanted to reduce the chances that readers would pick out these numbers without taking into account the discussion of their limitations.

Page 3952: A comparison with surface observations and a discussion have been added.

Page 3953: We've added that the OH value is the global-mean mass-weighted value, and now include the methane lifetime in the paper.

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We agree that our representation of isoprene chemistry is simplified, and this may contribute to biases, as we now note. However, since pretty much all tropospheric chemistry models seem to have this bias (to the best of our knowledge), we believe that it is unlikely to result from our simplified chemistry scheme.

Checking against another run (which differed only in its lightning parameterization, which generated more NO_x in the troposphere), we found that the NO_x flux across the tropopause was +0.6 Tg N per year in that run, as opposed to -0.6 in the run discussed here. It appears thus that this number is not terribly robust, even though it is a five year average, and that small changes in the circulation can have a large impact on this value. This is presumably the result of the net flux being a small difference between the much larger upward and downward fluxes. The net flux value is quite sensitive to the precise surface chosen (ozone was sensitive to this as well, as discussed in the text). Thus we are confident only that this is near zero, and have noted this in the table caption. Additionally, the programming error in the stratospheric ozone seasonal climatology had some effect on the stratospheric NO_x field, which will be corrected in future simulations. Since this flux is so small, we believe that this problem has not greatly affected our results.

As suggested, we've removed the model standard deviations, so all the plots follow a more uniform standard of symbols and quantities shown.

Page 3954: Another good point. We've added some comments on possible deficiencies in wet deposition to the text here. This is an important modeling issue which we've already begun working on further.

Page 3956: We've added the model's methane lifetime and a discussion of its initialization and the equilibration of its interhemispheric gradient. We thank the reviewer for bringing these oversights to our attention.

We also note that the interhemispheric gradient isn't purely a function of OH, as the reviewer points out, and that the Prinn et al data could perhaps be reconciled with the

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modeled OH given alternative exchange rates and/or source distributions.

Page 3957: We've clarified the sentence as suggested. We haven't added a budget table however, since as with the present-day simulation we hoped to reduce the chances that readers would pick out these numbers without taking into account the discussion of their limitations.

Page 3958: We now address the effect of PANs. Unfortunately, we did not save the tropopause diagnostics for these runs. It's an interesting point, however, and we've added them to our output for future simulations.

Page 3959: We now note that hydrocarbons produce HOx as degradation products. The reviewer's explanation of the OH shifts is excellent. This is what we were getting at, but we've revised the text to clarify this using some of the reviewer's suggestions. Oxidation of hydrocarbons can also produce HOx, but the shift in partitioning between HO₂ and OH is usually the larger effect, which we hadn't been clear enough about, so we focus on that now.

Lastly, we now note that we've included stratospheric temperature adjustment in the radiative forcing.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3939, 2003.

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