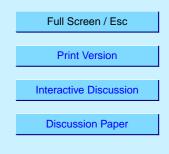**Atmospheric
Chemistry
and Physics
Discussions**

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 their comments, which have proved quite useful in revising the paper. In addition to many relatively minor comments, which we have addressed as detailed below, the reviewer raised the question of whether the particular simplified treatment of hydrocarbon chemistry that we used was appropriate over a wide range of conditions (a point also raised by the other reviewer). This was a very appropriate question, as our description of the chemical scheme was incomplete. In fact we chose to use this particular simplified chemistry precisely because we felt that it provided an excellent balance between a small enough computational expense to be suitable for climate simulations while having been extensively validated over a wide range of

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conditions and been shown to be appropriate for global studies. However, we neglected to explain the extension of the mechanism beyond the original CBM-4 model to make it useful for a broader range of conditions in our earlier manuscript. We are grateful that the reviewers noted this oversight, and we have now greatly enlarged the description of the chemical mechanismŠs formulation, as detailed below.

Specific comments

Page 3940 & 3941

The reviewer points out several sentences in the abstract and introduction which could be improved. We fully agree with all these suggestions, and have added to or clarified the text at the suggested locations.

Page 3942

As mentioned above, our description of the chemical mechanism, especially for hydrocarbon degradation, was inadequate. As the reviewer points out, the CBM-4 scheme was developed for polluted environments and would not necessarily be suitable for background conditions. However, the scheme we are using is a version 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). We now include this description in the text (section 2.1). While Houweling et al do not include acetone in their scheme, we will consider including it in the future to test whether it has s significant impact in the upper tropo-

sphere in our simulations.

We have added that XO2 (along with the other surrogate species) is kept in steady-state.

The Dentener and Crutzen paper provides both the uptake coefficient and the reaction coefficient, though we agree that the former is the main advance in that work.

We now note that clouds and aerosols both interact with the photolysis scheme.

Page 3944

The reviewer suggest several additions to the basic chemistry model description. We have therefore added the upper boundary of the chemistry calculation, the stratospheric climatology and the NOx from lightning, as suggested.

Page 3946

We have added the interhemispheric exchange time.

Page 3947

The reviewer raises a good point about equilibrium. We initialized long-lived methane to observed values, so it would equilibrate rapidly (starting off close to its equilibrium values), while other gases have much shorter lifetimes than our two year spin-up, which we now note.

Page 3950

We have toned down the qualitative portion of the discussion of the modelŠs ozone simulation. We now say that the model does a fairly good job based on the comparisons shown in Figure 2, and that itŠs certainly improved over the previous version, both of which seem to be fair statements. The quantitative comparison given in Table 3, however, is the most useful, and shows that the average difference is not hugely greater than the average bias (except for the 125 hPa level, where indeed the climatol-

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ogy showed poor seasonality due to a programming flaw in its implementation), which indicates that the seasonality is not wildly wrong.

The reviewer suggests that a longer discussion of surface ozone and a comparison with observations would be useful. We concur, and so have compared the model results with measurements from 40 stations, and added in the new Figure 4 showing a sample of 9 of those comparisons, along with a discussion. We thank the reviewer for this useful suggestion, and believe that the additional material greatly strengthens the model evaluation.

Page 3952

We examined the vertical profiles as the reviewer suggested, and note in the text now that they support this claim. Given the very large number of figures already included, we felt that it was better to state this in the text and not add another multi-panel figure.

Page 3953

We agree that the budget is useful for looking at the impact of stratospheric ozone on the troposphere, and state this in the text.

We note that the CO sites were chosen as in our previous paper simply to cover a wide range of latitudes and hence abundances. Also, we agree that the seasonality at Barrow and Cape Mears appears to be somewhat underestimated, which we now note, but we believe that overall the seasonality looks quite good.

We agree with the reviewerŠs comment that our simplified representation of hydrocarbons cannot fully capture all of isoprene chemistry, and now include this among the possible reasons for having the typical trouble when using a large emission inventory.

Page 3954

Two references to model overestimates of HNO3 were added.

Page 3955

WeŠve added that the lightning parameterization was adjusted by increasing the falsh per convective event empirical coefficients to give a better match to OTD data.

WeŠve revised the hydrogen peroxide discussion, toning down the commendation of our OH simulation and agreeing with the reviewer that our hypothesis that the H2O2 sink is the source of our model/measurement discrepancy is Śmost likelyŠ. WeŠve also noted that the model lacks uptake of HO2 on aerosols and in-cloud oxidation of SO2 by H2O2, which may also contribute to the positive bias, and that we plan to address both of these issues in future coupled chemistry-aerosol simulations.

Page 3956

WeŠve expanded the discussion of methane to give itŠs initialization, its lifetime, and to discuss how the interhemispheric gradient is calculated and how that timescale relates to the interhemispheric exchange time.

Page 3957

We now state that the surface types and the stratospheric ozone were both unchanged going to the preindustrial, which is useful information. We also discuss how the water vapor changed, though the emission changes dominate the ozone and OH response. Lastly, weŠve revised the test as suggested to clarify that weŠre discussing modeled ozone changes in this section.

Page 3964

The Derwent reference has been corrected.

Page 3969

As with our overall description of the hydrocarbon chemical mechanism, the description of the surrogate species (e.g. XO2) was inadequate. WeŠve expanded this to give a fuller explanation of what the surrogate species XO2, XO2N, RXPAR and ROR are. Again, we are grateful to the reviewer for pointing out this oversight.

Page 3972

Checking against another run (which differed only in its lightning parameterization, which generated more NOx in the troposphere), we found that the NOx flux across the tropopause was +0.6 Tg N per year in that run, as opposed to Ű0.6 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 a result of the net flux being a small difference between the much large 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 NOx field. This will be corrected in future simulations. Since this flux is so small, we believe that this problem has not greatly affected our results.

Page 3975

WeŠve made the use of line types and symbols uniform across the figures.

General

We were unable to expand the color scale and still discriminate readily between the colors. We prefer to stick to warm colors for positives, though this limits the range available, as we feel this convention is more easily understood.

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