**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.

Anonymous Referee #1

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This paper describes the latest generation of the GISS Chemistry-Climate model. Improvements on the previous model include the vertical resolution, boundary layer parameterization and the inclusion of simplified hydrocarbon degradation chemistry. Although the model does not present much new science, it does present the latest assessment by this model of climactically significant parameters such as the radiative impact of anthropogenically produced O_3. It is thus important and should be published.

My major concern is the usefulness of the hydrocarbon oxidation chemistry scheme that has been included in the model.

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It would be nice to see some comments about the model's simulation of H2O. Water vapor is important for tropospheric chemistry. The ability of the hydrological cycle in the model to respond to climatic perturbations in one of the advantages of this model and some discussion of how the model performs in this regard would be useful.

Specific comments

Page 3940

First sentence is rather long and uninspiring

"The chemistry now includes PAN and non-methane hydrocarbons in addition..." I think that this should read something like "The chemistry now includes a simplified representation of PAN and non-methane hydrocarbon chemistry"

"and contains a full representation of the stratosphere" The model contains a full representation of stratospheric dynamics, but contains no chemistry.

"Increased anthropogenic emissions of greenhouse gases and ozone precursor gases such as nitrogen oxides (NOx) produce tropospheric ozone" I'm confused over this statement, which greenhouse gases cause O3 production but aren't O3 precursors?

Page 3941

"Estimates are therefore based on model simulations, though these are subject to large uncertainties in emissions of ozone precursors in both modern and especially in preindustrial times". There are other uncertainties in the models than the emissions (chemistry, dynamics, physics). It may be worth mentioning them.

Page 3942

I am a little concerned about the choice of the updated chemistry scheme. The carbon bond scheme was developed for computationally rapid modeling of local and regional photochemical smog. In these condition NOx concentrations are high, however for most of the global atmosphere NOx concentrations are much low. The authors should

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specifically acknowledge this and provide specific evidence that they (or others) have investigated the restrictions that this places upon their calculations.

I suspect that for the scientific uses that authors have outlined, the chemistry scheme will not be completely suitable. The authors are especially interested in the upper troposphere; however they can have little confidence in the HOx budget in that region as they are missing the required acetone chemistry. Cross-peroxy radical reactions (RO2+RO2) are not fully represented in the scheme (notably reactions between CH3O2 and the higher peroxy radicals) which may provide problems in the cleanest atmosphere.

Whilst I agree that there is great value in having a model that can be run quickly or for extended periods of time I think that there is also value in having confidence in the chemistry scheme that you are using. The authors should provide more evidence that this scheme is suitable for the simulations they want to do.

Is XO2 also kept in steady state along with HOx, CH3O2, C2O3?

Doesn't the Dentener and Crutzen paper give a reaction probability (gamma) for the uptake rather than a rate constant.

Do the clouds and aerosols interact with the photolysis scheme?

Page 3944

How is the tropopause defined from the perspective of running the chemistry and forcing the stratosphere?

Is there any stratospheric loss of CH4, CO, etc?

Can the mass of NOx from lightning be mentioned in this section? I realize that it is not a completely prescribed field but it would be useful to be able to compare it to the other sources.

Page 3946

The inter-hemispheric mixing time should be specifically given.

Page 3957

"The model reaches equilibrium with the first year" How is this defined? Presumably there are species (such as to CH4) which the model isn't in equilibrium for.

Page 3950

"In general the model does a good job of reproducing both the magnitude and the seasonality of tropospheric ozone." I'm not sure what a "good job" means in this context. The authors have shown that the new model's simulation of O3 is an improvement over the previous version. They have not really discussed the seasonality of the O3 nor shown that the model has much skill in this regard. At most altitudes and locations in figure 2 the model maxima and minima O3 concentration fails to be within the same month as the measured maxima / minima.

The comparison with surface O3 is also weak. The section is too short to be particularly useful in offering a constraint on model processes. A longer section that showed observed and simulated surface concentrations would be useful.

Page 3952

"If the dry deposition and chemistry were ..." Although there may be a reasonable point here the authors should show some vertical profiles of O3 to show that the gradient at the surface is well represented.

Page 3953

I agree with the authors that O3 budgets can cover up a multitude of sins occurring within the model. However, they are important to asses the role of tropospheric vs. stratospheric influence on the O3 concentration.

Why were these CO sites chosen specifically?

The authors seem particularly happy with their OH simulation. However it appears from Barrow and Cape Mears that the seasonality is under represented by the model.

"... understanding of NHMC chemistry" I donŠt think we can expect the scheme used here to represent all facets of NHMC chemistry especially for something as complex as isoprene.

Page 3954

A reference should be given for the general model overestimate of HNO3.

Page 3955

"empirical adjustments made to the parameterization" What are these adjustments?

"Given the overall high quality of the OH-simulation" Again I think that this might be an overtly strong commendation of the model. The methyl-chloroform lifetime is within an accepted range, however nearly all of this is occurring within the tropical boundary layer. I donŠt think it provides much information about the geographical distribution of OH.

There is also a certain degree of luck involved here. The model does miss some tropospheric photochemistry (uptake of HO2 on aerosols and change in N2+O(1D) rate constant for example) so any agreement has to be viewed with a certain sceptism.

However, I agree that the most likely explanation of the model deviation from the measurements of H2O2 is the sink term. As Asia is a large SO2 emitter it may be the oxidation of SO2 by the H2O2 in clouds.

Page 3956

What is the methane initial condition? Presumably the model isn't in steady state after a 7 year run.

Page 3957

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Do the dry-deposition surface types change in the pre-industrial case? What about the stratospheric climatology?

How has the H2O concentrations changed in the model?

"Tropospheric ozone levels have increased ..." The text should emphasis that this is the modeled O3 concentrations.

Page 3964

Derwent reference should be volume 24 not 30

Page 3969

XO2 is described as a NO to NO2 operator. Is this different to a general organic peroxy radical? Similarly for XO2N. What is a paraffin budget corrector?

Page 3972

Why is the stratosphere behaving like a sink for NOx? Most models have a small NOx source from the stratosphere.

Page 3975

Can the usage of triangles, squares and lines for the different model runs remain the same between figures?

General

I don't like the blockiness of the color scales. More colors would help show some features - especially how low the O3 concentration gets to in figure 4.

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