

***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.**

**W. Collins (Referee)**

bill.collins@metoffice.com

Received and published: 31 July 2003

This work is a useful update on a previous paper (Shindell et al. 2001). The model has been substantially improved in this work through the addition of non-methane hydrocarbon, and an increased vertical resolution. Both of these additions are essential for modelling the radiative forcing from ozone. The higher hydrocarbons enhance the transport of NO<sub>x</sub> (as PAN) and HO<sub>x</sub> (as oxygenated hydrocarbons) out of their source regions, and the increased vertical resolution is necessary to model ozone around the tropopause where it has the greatest radiative effect. Even though the forcings calculated here are very similar to the Shindell et al. 2001 paper they are much more

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

credible, and hence much more useful to the community.

I was interested by the authors remarks concerning ozone global net chemical production. Great importance is often given to this number, but I agree with the authors that it is actually of little use.

My only major criticism is that surface ozone concentrations are discussed at length, but the only comparisons are with a few sonde data. To validate the surface concentrations comparisons need to be made with a number of surface sites (such as from the Logan compilation) rather than sondes. I am suprised at the low concentrations simulated over Africa and South America.

I recommend that this paper be published with an additional figure of surface ozone comparisons, and with minor revisions detailed below.

Detailed comments: Page 3941, line 20: Hauglustaine and Brasseur 2001, JGR 106 p32337 would also be a good reference to quote here.

Page 3943, line 8: Has the scheme been tested in remote conditions such as the low NO<sub>x</sub> high VOC of the tropics, or low NO<sub>x</sub> low VOC of the free troposphere?

Page 3949, line 8: I assume the sonde measurements quoted for 950 hPa are actually the averages of those at 1000 and 900 hPa. This should be made clear. Three of the sites do not have 1000 hPa measurements, hence the plots are for 900 hPa, not 950.

Page 3948, line 20: I am rather worried by the very poor simulations in the northern hemisphere at 125 hPa and 200 hPa, particularly the failure to represent the seasonal cycle. These points are either in the stratosphere or dominated by advection from the stratosphere and hence should be easy to simulate. The authors attribute this to "biases in the stratospheric ozone climatology". These biases may be sufficiently large to distort the radiative forcing calculations. Have the authors considered using a more realistic climatology? Is it possible that some error has occured when prescribing the climatology?

Page 3949, line 7: Figure 2 does not show surface measurements, but an average of sonde measurements at 1000 and 900 hPa. Hence they are not really suitable for assessing boundary layer and dry deposition schemes. The model results at 950 hPa will not be from the surface layer either, but from layer 2 (945 hPa). Comparisons with surface sites are needed.

Page 3949, line 15: While the mean biases may now be small, that is the result of averaging large positive and negative errors.

Page 3950, line 19: How do the N. Atlantic data compare with Izana observations?

Page 3950, line 26: I assume "2 ppbv" should be "20 ppbv".

Page 3951, line 3: I assume "hydrocarbons" haven't actually increased in the new version since they were emitted as CO in the previous one. However alkenes (especially isoprene) can react with ozone whereas CO can't. There may also be a large effect due to changes in the dry deposition (of both ozone and NO<sub>2</sub>). The destruction over the tropical forests may now be too large.

Page 3951, line 6: I suspect that the ozone budget is only "substantially altered" when looking at the net production which is the difference of two large terms. It is the gross production and destruction that are important, not their difference.

Page 3951, line 10: It is not surprising that changes in strat-trop exchange are compensated for by the chemistry. Ozone is approximately in steady-state in the troposphere so any decrease in STE necessarily implies a compensating decrease in destruction. The gross production of ozone in the troposphere is about 3000-4000 Tg/yr (Collins et al. 200 QJRMS 106 p1925) so a change in STE by 364 Tg/yr would only effect the burden by 10%.

Page 3951, line 22: Similarly this is not surprising. To balance the budgets, any increase in net production must be met by an equivalent decrease in dry deposition. An increase of 201 Tg/yr is approximately 5% of the gross production, hence the burden should

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

increase by about 5% - which it does.

Page 3951, line 26: It would be clearer to present these numbers in a table.

Page 3952, line 23: I'm not convinced that the authors have shown that the surface concentrations are well simulated, only two of the stations in figure 2 are continental sites (Hohenpeissenberg and Natal). The authors should compare with a selection of the Logan surface sites.

Page 3953, line 11. Over what domain is the OH abundance averaged? Global mean OH cannot be directly compared with the MCF lifetime (Lawrence et al. 2001 ACP 1 37-49). The authors should compare like with like, eg. methane lifetimes.

Page 3953, line 21: The biases might not be due to "deficiencies in current understanding of NMHC chemistry", but to deficiencies in the implementation of isoprene in this lumped chemistry scheme. Such deficiencies are the lack of representation of the isoprene degradation products (e.g. MVK, methacrolein, methylglyoxal and their hydroperoxides) which are readily removed by wet and dry deposition.

Page 3953, line 24: I'm suprised that the stratosphere is a net sink of NO<sub>x</sub> in table 4, Murphy and Fahey 1994 suggest a source of 0.45 Tg/yr.

Page 3953, line 25: In figure 5 the model standard deviations are orders of magnitude smaller than the observations. I assume they are calculated differently, in which case they should be omitted as they do not add anything.

Page 3954, line 22: Could deficiencies in the wet deposition scheme be an explanation? The authors refer to this later in the H<sub>2</sub>O<sub>2</sub> comparisons.

Page 3956, line 11: What is the modelled methane lifetime? How is the methane initialised? Is the interhemispheric gradient a result of the initialisation or has it reached equilibrium?

Page 3956, line 20: The interhemispheric gradient is not simply a function of the OH,

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)

the interhemispheric exchange rate and distribution of the methane sources are all important. It would probably be possible to get a reasonable interhemispheric gradient with the Prinn OH if the exchange rate and source distributions were tuned within their uncertainty ranges.

Page 3957, line 19: "Tropospheric"->"Modelled tropospheric"

Page 3957, lines 22-27: These numbers would be clearer in a table.

Page 3958, line 11: Should mention the effect of PAN here.

Page 3958, line 15: Did the tropopause change height between the simulations?

Page 3959, line 1: How do increased hydrocarbons lead to HOx production?

Page 3959, lines 5-10: Surely the explanation here is that increased NOx pushes the OH-HO2 balance towards OH, increased hydrocarbons push the OH-HO2 balance towards HO2. The lifetime of NOx is much shorter than that of hydrocarbons, so NOx dominates near the source regions, hydrocarbons dominate in the free troposphere.

Page 3959, line 14: Have the authors included a stratospheric temperature adjustment (IPCC TAR, chapter 6)?

---

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

[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)[Discussion Paper](#)