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Interactive comment on "The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990–2030" *by* F. Dentener et al.

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In addition to our previous general comments, we reply here to the detailed comments of reviewer #3.

The manuscript has been somewhat re-organized to accommodate the remarks of the reviewers, as well as informal comments of readers. Most notably: we have re-organized the emission section so that parts of the discussion have been moved to section 2; and -for the interested reader - to the appendices. We have moved the discussion on radiative forcing from the discussion section to section 6. We have reduced

the amount of surface ozone plots. We refer in the text to the original ACPD paper for the NH summer plots. We have performed some additional studies to convince ourselves and the reviewers of the sensitivity of certain processes (ships and NMVOCs).

1.S3310; in abstract. “and would reduce the radiative forcing of ozone and methane between the 1990s and the 2020s to approximately -0.1 Wm-2. This can be compared to the 0.14-0.47 Wm-2 increase of methane and ozone radiative forcings associated with the SRES scenarios.”

2.S3310 l. 22. It is correct that most model studies predict a small decrease or increase of OH going from the pre-industrial to present day atmosphere [e.g. Wang and Jacob (1998); Lelieveld et al. (2004); and a multitude of earlier references therein.] The interesting outcome of this study is that ‘only’ abating air pollution or CH4, may have a stronger impact on OH. We added a sentence at the end of section 5.1

3.S3311 l. 33; We toned down our statement that our models ‘realistically’ reproduce measured trends; however we retain ‘a broad agreement’ of models and measurements. We leave the discussion of the match with CO and CH4 trends at the end of section 4.2.3 and 4.2.4. We did not compare modeled ozone trends with measured ones, however as can be seen from the new Figure 5; there are for the time period considered no obvious strong trends in surface ozone. Further we improved the quality of Figure 7 (new Figure 4) to improve readability (and found an interpolation error improving STOCHEM results). We admit that our ‘worse’ case comparisons of models and measurements at continental stations are of the order of 10-15 ppbv. On request of the reviewer we also add correlation coefficients (on the basis of monthly mean values between 1991-2002) and mean values. Given the relatively coarse resolution of our two models (horizontal resolutions of 400-800 km), we think we should not be too demanding on reproducing continental scale O3 concentrations, especially not in regions with high emissions density; as mentioned in the discussion section. The capability of global models to reproduce surface ozone in various world regions is the topic of on-going work in the ACCENT-IPCC-AR4 context. Initial analysis tells that global mod-

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els often show O3 differences exceeding 10 ppbv at various world locations. Whether these models can be called realistic or not, is a matter of definition.

4.S3312 l. 9 A spin-up of spin-up of 13 months for STOCHEM and 24 months for TM3 was used (section 4.1). Due to unforeseen inaccuracies in the initialization, TM3 CH4 is about 30-40 ppbv lower than the measurements, whereas STOCHEM is about 30 ppbv higher (section 4.2.4). Although we discovered these inaccuracies only after finalizing the 40 years simulations- we argue that the results do not critically depend on it.

5.S3312 l. 13 We performed an additional sensitivity study showing that 1-1.4 ppbv O3 increases are due to ship emissions (see section 4.2)

6.S3312 l. 15 We deleted former Figures 12b/c

7.S3312 l. 21 We recognized the repetition. We re-organized the emission section such that the main points are made in Section 2; and the comparison with other work is made in the Appendix. Note also that in Cofala et al. (2005), available on the web, further details are presented.

8.S3312 l. 23 Aerosol interactions with radiation and photolysis rates were not included in our models; and e.g. in India and China it should have some impact on surface ozone. A recent study focusing on this by Bian et al. [JGR, 2003] revealed impacts on the order of a few ppbv. The impact of coincident aerosol emission changes on aerosol (and ozone) in e.g. India, is a topic that goes beyond the scope of our paper, but may be of similar magnitude. We thank the reviewer for pointing out this interesting interaction - which we certainly will pick up in future work.

9.S3311 l. 25 The stratospheric fluxes were 520 Tg for TM3 and 420 Tg for STOCHEM (through 100 hPa level), both with little (10-15 Tg/yr, plus minus 1 standard deviation) interannual variability. These numbers are also given in the model description

10.S3312 l. 1 Indeed to make our models fully comparable we decided to calculate

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all budgets up to 100 hPa. This includes a fair bit of stratospheric ozone in the extra-tropics. When only the troposphere is included our models have tropospheric burdens of about 270-330 Tg O3; as given in (Stevenson et al., 2004; Table 2) and the IPCC TAR report. This is mentioned in section 5.1

11.S3312 l. 5; we have analyzed the stratospheric contribution to surface ozone in TM3 according to the method described by Lelieveld and Dentener (2000); above the Himalaya it amounts to 25 % of the annual average (see text), elsewhere generally less than 10 % (annual average).

12.S3312 l. 9 we have added a sentence in section 5.2 on how biomass burning emissions are included. "The biomass burning signal in tropical Africa is much stronger in STOCHEM than in TM3, probably due to differences in the mixing schemes of the two models, or in the different geographical and temporal distributions of biomass burning emissions in our models. It is likely that biomass burning emissions are partly released at higher altitudes, leading to more effective ozone production, as implicitly included in STOCHEM, but not in TM3."

13. S3312 l.11 In the abstract, discussion and conclusion we discuss the difference between CLE and BAU, and that we expect that by necessity legislation will be increasingly followed.

14. S3312; l.13 In Section 6, we mention that indeed the increases are found throughout the troposphere.

15. S3312-S3314. Almost all very detailed remarks were very helpful and included in the manuscript. Not included were:

16.S3314, l.6 Table 1 left as was; emission totals can be read from Figure 2-4.

17.Table 2; Altitude information was added, the 2030 burdens can be found by combining new Figure 9 and Table 2.

We thank reviewer #3 for the detailed comments which helped to improve the

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manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 8471, 2004.