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Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #3

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General Comments:

This paper makes several important contributions, meriting its publication in ACP. First, the authors present two new near-term (1990-2030) projections of ozone precursor emissions, one that accounts for regulations on ozone precursors that are already being legislated (Current Legislation (CLE) scenario), and one that explores emission controls that could be implemented with available technology (Maximum Feasible Reduction (MFR) scenario). These scenarios are more optimistic than the SRES sce-

narios considered in the 2001 IPCC report. The authors then use two different atmospheric chemistry models (the Lagrangian STOCHEM and the Eulerian TM3) to quantify the impacts of the CLE and MFR scenarios on atmospheric composition, with a focus on implications for climate and ozone pollution. Their findings support earlier work demonstrating that methane control offers the potential to mitigate both climate change (through decreased radiative forcing from both methane and ozone) and air pollution (through decreased surface ozone levels). Before publication in ACP, there are several points that should be clarified (outlined below), as well as some editorial changes to improve readability.

Specific Comments:

Abstract & conclusions: A comparison of the radiative forcing and ozone concentrations predicted with the CLE and MFR scenarios versus those from IPCC SRES would emphasize the optimistic nature of the new scenarios.

Decreases in CO and NMVOC should increase OH and decrease the methane lifetime. Thus, the results presented in the abstract ("further reductions of air pollutants NOx and NMVOC result in lower ozone, but increase lifetime of methane") and on p.19 line 21 ("OH is reduced as a consequence of the lower NOx, CO, and NMVOC emissions") reflect the sensitivity of the global atmosphere to changes in NOx emissions as opposed to the combined influence of CO, NMVOC, and NOx. This compensation between the influence of NOx and VOC + CO on OH may be what the authors intend to convey (on p. 20 lines 4-8) when they state that CLE and MFR are "balanced scenarios", but should be clarified. This balancing is consistent with modeling results from Wang and Jacob [1998; J. Geophys. Res., 103, 31,123-31,135] who found that OH only decreased slightly from pre-industrial to present-day despite increases in emissions.

There are phrases throughout the text (abstract: "reproduce realistically"; p. 14, "safely used to extrapolate", top of p. 28, again in conclusions) asserting that the models reproduce observations. Some quantitative justification, such as a correlation coefficient

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or bias estimate would strengthen these statements. Figure 7 in particular is too difficult to read to make a convincing conclusion, and the 10-15 ppbv ozone discrepancies over continental stations (p. 14, line 20) seem to contradict assertions of a realistic simulation. Matching the observed trends is clearly beyond the scope of this study but the authors may wish to remind the reader that inter-annual variability in meteorology undoubtedly contributes to the observed concentrations but is not included in the simulations, which were designed to isolate impacts of changes in anthropogenic emissions on atmospheric composition.

p. 17 line 6: "inaccuracies in the initialization done differently in both models." The methane initialization should be explicitly described since it is relevant to the results. Discussion of the long methane lifetime and implications for the transient experiments (particularly as compared to the shorter-lived air pollutants) could be included.

Is the maximum ozone (e.g. Fig 12) over the oceans associated with ship emissions or with coastal surface emissions trapped in ocean boxes with shallow mixing depths?

Including both Figures 12 and 13 seems repetitive. I recommend showing Fig 12a but cutting 12bc since, as the authors point out, their main points are made with Fig 13.

Section 2 suffers from some repetition, which might be avoided by combining Figs 1-3 and discussing them together. The treatment of NMVOC should be discussed earlier. Section 7.1 provides a good synthesis of the emissions scenarios and a useful comparison with existing inventories and projections. I recommend moving most of this discussion, to Section 2, to provide the overview for the reader (of how these new emissions compare with existing inventories) before presenting the model results.

Section 3 (Model Description): –Are aerosols included in the models and allowed to interact with chemistry and photolysis rates? Neglecting such interactions is probably most relevant for the large simulated ozone increases over India. –How different are the annual mean cross-tropopause fluxes of ozone in the models? –How much STE inter-annual variability does STOCHEM exhibit (p. 18 lines 14-15)

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p. 19: The present-day ozone burden of 450 Tg seems fairly high compared with other models (e.g. IPCC 2001 Chapter 4). Is this due to the inclusion of the stratosphere in the burden calculation? Why not use a lower tropopause for the mid and high latitudes to more accurately represent the tropospheric burden?

p.20: "high concentrations over the Himalayas are due to strong mixing with stratospheric air." Was this explicitly diagnosed with the model? Free tropospheric ozone concentrations are generally higher than at the surface (away from source regions) and are not necessarily associated with stratospheric air.

p. 20 last line: differences in the implementation of biomass burning are mentioned but not described. Discussion should be included when the models are presented.

p.21 CLE results: The authors may wish to emphasize that increases are occurring despite implementation of currently proposed emission controls.

p.22, lines 26-27: Peak values coinciding with elevated surface concentrations. Ozone radiative forcing maximizes in the upper troposphere, so this result implies that concentrations are elevated throughout the tropospheric column. This could be explained.

Technical Corrections:

p.4 lines 14-15 "proposed to increase" is awkward

p. 4 line 16, "compared to present background levels of 30-35 ppbv" is this range based upon observations [e.g. Vingarzen ref] or models [Prather ref]?

p.5 lines 10-11: delete extra "and ozone"

p.6 line 25: "Similarly scenarios" sentence is redundant given statement in line 21.

p.7 lines 7-8. Awkward phrasing; are Asian emissions growing by 35% from present?

p.9 line 17: delete related. Phrasing is awkward; is the figure showing uncertainty?

p. 9 aircraft emissions: why was a polynomial fit chosen?

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Interactive Comment

p.10 line 19 "quite high emissions": compared to other inventories or other regions?

p. 11, lines 1-2: "hard-wired into the model code" is unclear; does this mean that they vary with the meteorology used in the model?

p.15 lines 8-9 "interpolated to station location..": why was geographic interpolation necessary rather than sampling the appropriate model box?

p.15 line 16: delete "slightly"

p.19 lines 1-5: needs rephrasing

pp. 19-20: combine discussion of CH4 lifetime with CH4 burden to avoid repetition

p. 20. lines 10-13 CH4-OH feedback could be explained more clearly, emphasizing that increases in CH4 concentrations reduce OH, thereby lengthening the CH4 lifetime, and causing atmospheric CH4 concentrations to rise further.

p.20 section 5.2: explain 1990s and 2020s (i.e., mean 1990-2000?)

p. 21, line 2 "ozone background": the 50-60 ppbv concentrations are not in particularly remote locations (India, downwind of U.S.) so background seems inappropriate.

p. 21 lines 16-17 "global nature of ozone control strategiesis": unclear, perhaps the authors intend to say, "illustrate the need for global ozone control strategies"?

p. 21 line 17 "non-linear NOx NMVOC chemistry": please explain further.

p. 21 lines 20-21 "ozone-NOx titration dependent on coarse model resolution." Shouldn't there be more titration in a finer resolution model with concentrated sources?

p. 21 line 9: "growing season" is often longer than June-August.

p. 22 line 15: is the SH strongly affected during its growing season?

p. 28 line 9: "overcompensating" should be "offsetting"

p. 28 line 13: "as in future" should be "and in the future"

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p.34 line 13 typo- "sparse" instead of "sparser"

p.34 line 16: typo- extra parenthesis

–Many IIASA internal reports are cited (e.g. Cofala 2004b, Mechler, 2003) . If they are available on the web, please cite the address.

–Akimoto reference is missing

–Table 1: Meteorology column can be deleted since it is in the text. Table could include base 1990 and 2030 global emissions for NOx, CO, NMVOC & CH4 in both models for each simulation rather than requiring readers to estimate from Figs 1-3.

–Table 2: Include altitude for burden calculation. While Figure 11 shows the future trends, the normalization to 2000 masks how model-to-model differences compare to differences among the scenarios. Including the global burdens for the 2030 scenarios in the table would enable this comparison.

–Table 3: What is meant by "Other: emissions of NOx-CO-NMVOC" in the table title?

In general, all Figures would benefit from larger text. Figs 1-3 and Figs 4-6 could be combined into 3-panels-per-page to ease comparison.

Figure 10 caption: state if the model was sampled only at the surface NOAA stations, or throughout the domain. The label MRF should be MFR in the figure.

Fig 5 caption: explain why CO from biomass burning is unrealistically high.

Fig 11 caption: state that TM3 is solid lines; STOCHEM dashed.

Fig 11: either add (a), (b) to figure panels or remove from caption.

Fig 13 caption: state that we are looking at 2020s-1990s.

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