Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of high solar zenith angles on dynamical and chemical processes in a coupled chemistry-climate model" *by* D. Lamago et al.

Anonymous Referee #1

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Overall this article is an important contribution to the field of stratospheric chemistry. It shows that twilight conditions should not be neglected in stratospheric chemistry models because twilight photolysis has a clear impact on chemistry and consequently on dynamics .

The paper is clearly organized and written and presents most of the results adequately. However, it is very brief in some points, see below, and it would further improve the manuscript explaining these points in more detail. After taking into account these points the manuscript will be acceptable for publication in ACP.

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Major points:

1. The parameterization of photolysis frequencies:
   This is the key improvement of the E39/C model discussed in this paper. There-fore, this improvement needs to be described in enough detail, which is not the case: the Landgraf and Crutzen model is used for $\theta \leq 87.5°$ and a variant of the Röth parameterization for $\theta > 87.5°$. How the extension for $\theta > 87.5°$ has been done is not clear. Which reference photolysis frequencies have been used for the fitting procedure? The Röth parameterization involves the two parameters a,b whereas in Eq. (8) there is only b_i; what is the impact of this simplification?
   Further, Eq. (9) gives the air mass factor for $\theta < 90°$; however, it is not explained how $d(\theta)$ is calculated for $90°\theta < 93°$.
   Finally, it is not demonstrated in the manuscript how well the parameterization works. It would be helpful to show a comparison of the parameterized photolysis frequencies with the more precise reference frequencies used for deducing the b_i parameters at least for a few key species.

2. No comparison with observations is made in the paper as it stands. I suggest to add some discussion on this issue, because otherwise there remain open ques-tions:

   - How realistic are the simulated ozone fields?
   - From Fig. 11 & 13 one concludes that chlorine deactivation starts in mid October –is this realistic? Compare with e.g. Grooß et al. (1997).
   - In the 87.5 runs the chlorine deactivation in the Antarctic seems to happen via conversion of ClO_x to $ClONO_2$. It should be more clearly stated that such a model behaviour is unrealistic (Douglass et al., 1995; Grooß et al., 1997) and that therefore the SZA93 runs produce more reliable results.

3. Page 3, section 3.2.1, first paragraph:
   The authors write that '.. a closer inspection of Figures 3 and 4 shows ... the ozone hole appears approximately 2 weeks earlier than in the SZA87.5 run .. the lifetime ... is about one week longer ...'
   The earlier appearance and longer lifetime of the ozone hole is an important result of the study, but can not clearly be found in these Figures by closer inspection. An extra Figure zooming out this result should be presented, for example a plot of ozone (SZA87.5 and SZA93) versus time at $70°S$ and 50hPa.

4. Lary and Pyle (JAC, 1991) have shown that including zenith angles greater than $90°$ improves markedly their simulations of midlatitude NO_x. Does this effect likewise appear in the CCM results presented in the manuscript? Could a different NO_x impact midlatitude ozone colums?

5. Page 3, section 3.2.1, second paragraph:
   The authors write 'Rex et al. (2003) showed, that photochemical box model calculations cannot fit observed ozone loss rates..'.
   There are earlier references showing this feature, some of them should perhaps also be cited here (e.g. Hansen and Chipperfield, 1997; Becker et al.; 1998). More importantly, it should be made clear that the discrepancies between models and observations reported by Rex et al. (2003) and in other studies cannot be due to the models restricting the photolysis calculations to solar zenith angles of less than 87.5.

6. Page 4, section 3.4, first paragraph:
   '... employing equivalent potential vorticity coordinates...'
   should be better explained. What is equivalent PV? Link to equivalent latitude?

7. Page 5, section 3.4, last paragraph:
   '... Reduced $ClONO_2$ mixing ratios are found in the SZA93 run inside the polar

vortex ... while HCl is increased there...'
The explanation of this behaviour is confusing and hard to follow, please rewrite.

8. Figure 5: There is a statistically significant change in total ozone north of say 50°S that cannot be directly due to ClO-dimer photolysis. There should be some discussion of this issue in the paper. Further, the second and fifth sentence in the conclusions seems to be in contradiction with the results shown in Fig. 5: The effect of the SZA93 run is noticeable throughout the year and in certain month up to the equator.

9. Figure 6: It is not obvious what this Figure means, please explain clearer. For example, the standard deviation with respect to what average is meant? Is this Figure really necessary?

Minor points:

1. I suggest to change the title somewhat: 'high solar zenith angles' is confusing. Usually one does not talk about 'high angles'. 'High solar elevation' is a proper term (but high solar elevation is of course connected with small solar zenith angles). Further, all the discussion in the paper is on stratospheric issues so that it would be helpful to convey this piece of information in the title. Perhaps:

   Impact of LARGE/TWILIGHT solar zenith angles on dynamical and chemical processes in a coupled chemistry-climate STRATOSPHERIC model

2. Below eq. (9): replace 'this formula' by '$d(\theta)$'

3. Sec. 2.3, last two sentences: where are the mixing ratios of CO_2 etc. fixed? At the lower boundary? See also Table 2.

4. page 5, left hand column: replace 'chlorine products' by 'chlorine compounds'

5. Conclusion, section starting with: In the SZA93 run... Are these two sentences not in contradiction? Is the strongest effect at the edge or at the south pole?

6. Fig. 1: How is polar night defined here? Which is the SZA chosen as the cut-off?

7. Some typographical isues need improvement:

   - Chemical symbols should be in roman characters even if used in equations. E.g. use

   ```
   \begin{equation}
    \mathrm{CH}_4 + \mathrm{Cl}  \rightarrow  \mathrm{HCl} +
     \mathrm{CH}_3
   \end{equation}
   ```

   to obtain

   $$CH_4 + Cl \rightarrow HCl + CH_3 \tag{1}$$

   - Further, in equations functions should be in roman characters (and not in italics) E.g. use

   ```
   \begin{equation}
    \sin(x)
   \end{equation}
   ```

   to obtain

   $$\sin(x) \tag{2}$$

   and avoid

   $$sin(x) \tag{3}$$

   - Finally, units in the text should not be in italics.

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