Interactive comment on "Atmospheric changes caused by galactic cosmic rays over the period 1960-2010"
by C. H. Jackman et al.

Reply to Referee #2
We thank Referee #2 for helpful comments and suggestions. The "Referee's Comments" are noted first and then we give our "Reply:" to the comment.

Referee 2: **Anonymous Referee #2**
Received and published: 29 December 2015

Referee #2 *General comments: The manuscript describes the atmospheric changes caused by the influence of Galactic Cosmic Rays (GCR) on the production of nitrogen and hydrogen oxides. The authors analyze the response of the chemical composition to GCR simulated with two global chemical models using several scenarios of the boundary conditions. The subject of the manuscript is relevant to the ACP scope. The paper is well written and structured. The reference list includes most of the previous publications on this subject. All figures and tables are of good quality. The results contains already known information about the influence of GCR on the chemical composition as well as some new results concerning the influence of atmospheric state (e.g., chlorine and stratospheric aerosol loading) on the global mean total column ozone response to GCR. I think the publication of the manuscript can be recommended. However, there are several issues in the manuscript (see specific) comments and some moderate revisions would be necessary before the publication.*

Specific comments:
Referee #2 *- 1. The response of the chemical composition to GCR obtained with the exploited models agrees well with the results published by Calisto et al. (2011), however it is heavily underestimated in comparison with the results of Semeniuk et al. (2011). This disagreement was briefly discussed by Mironiova et al. (2015, 10.1007/s11214-015-0185-4) and I think it should be also discussed in the paper, because it is important for the community.*

Authors' Reply to 1.: We were not aware of the review paper by Mironova et al. (2015) before, thus did not reference it in our original manuscript. The results of CMAM given in Semeniuk et al. (2011) are discussed in Mironova et al. (2015), who note on pp. 59-60 of their paper that:

"CMAM results reveal statistically significant NO_x increase in the entire troposphere/lower stratosphere reaching up to 100 %. The reasons for such a substantial disagreement between the simulated NO_x responses are not clear yet. It may be explained by different background NO_x fields in the troposphere and lower stratosphere. The absence of anthropogenic and natural NO_x emissions together with oversimplified tropospheric chemistry in CMAM (Semeniuk et al. 2011) could lead to very small background NO_x abundance and strong impact of the GCR induced source. This hypothesis is partially supported by closer agreement in the troposphere over the southern high latitudes, where the influence of anthropogenic and natural sources of NO_x is the smallest.

The effectiveness of the ozone production by additional NO*x* strongly depends on the background NO*x* field. In the NO*x*-poor environment the ozone production can be very large, while for the relatively high level of NO*x* the ozone production by additional NO*x* is limited. Presumably the low background NO*x* mixing ratio in CMAM model is the reason of the large (up to 15 %) ozone enhancement in the entire troposphere, while in the CCM SOCOL significant ozone response is confined to the relatively clean southern hemisphere and reaches only 2–3 %."

It is outside the scope of this manuscript to discuss in great detail the much larger GCR-caused atmospheric changes in Semeniuk et al. (2011) compared to those given in Calisto et al. (2011) and presented here. Section 5.1 of the paper is slightly modified to note this difference in the results of previous papers.

Authors' modification of Paper as a result of 1.: We do now mention the much larger response of the CMAM model to the GCR perturbation at the end of the first paragraph in section 5.1. We add these two sentences:

As an aside, the SD-WACCM results, like those in Calisto et al. (2011), indicate a much smaller GCR-caused NOx impact than computed in Semeniuk et al. (2011). Mironova et al. (2015) propose that "the absence of anthropogenic and natural NOx emissions together with oversimplified tropospheric chemistry in CMAM" may be the reason for the larger response of the GCR perturbation in CMAM.

**Add Mironova et al. (2015) to the Reference list:**

Mironova, I. A., Aplin, K. L., Arnold, F., Bazilevskaya, G. A., Harrison, R. G., Krivolutsky, A. A., Nicoll, K. A., Rozanov, E. V., Turunen, E., Usoskin, I. G., Energetic Particle Influence on the Earth's Atmosphere, Space Sci. Rev., 194, 1–96, 2015

Referee #2 - 2. Comparison of the main results against Calisto et al. (2011) and Semeniuk et al. (2011) requires some comparison of the applied ionization rates, because the difference between NAIRAS and Usoskin et al. (2010) calculations should be well characterized.

Authors' Reply to 2.: The NAIRAS ionization rates from Mertens et al. (2013) are compared with those given in Usoskin et al. (2010) for solar minimum (1965) and solar maximum (1960) conditions in new Figure 3 in the paper now. The Mertens et al. (2013) rates are about a factor of two smaller than those given in Usoskin et al. (2010).

Authors' modification of Paper as a result of 2.: This is now discussed in the revised manuscript in the fourth paragraph of Section 2, which reads:

The Mertens et al. (2013) GPIR are about a factor of two smaller than those presented in Usoskin et al. (2010), and the altitude of the maximum in the GPIR is lower in the NAIRAS results as well,   A comparison of these two computations of GCR ion rates at 90 degrees N is given in Figure 3 for both solar minimum (1965) and solar maximum (1960) conditions. The underprediction of the NAIRAS GPIR and the lower altitude of its maximum is due to the lack of pion-initiated electromagnetic cascade processes in the HZETRN version 2010 currently implemented in the NAIRAS model (Mertens et al., 2013). This deficiency will soon be rectified when the 2015 version of HZETRN is integrated into the NAIRAS model (e.g., Norman et al., 2012, 2013; Slaba et al. 2013).
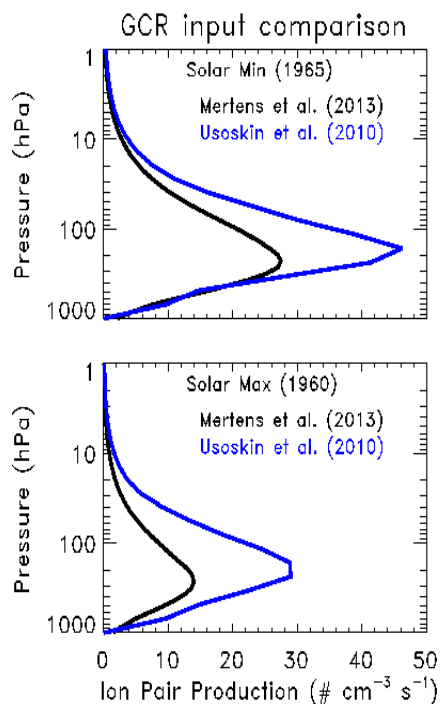
New Figure 3 caption:

      NAIRAS model computed galactic cosmic ray annual average ionization rates (Mertens et al., 2013) compared to those given in Usoskin et al. (2010) for solar minimum (1965, top plot) and solar maximum (1960, bottom plot).

Referee #2 - *3. The choice of the models is not justified. I do not understand why the models with prescribed dynamics/transport were chosen. If some influence of GCR on ozone concentration is expected than this model choice hampers the possibility to study subsequent effects of GCR on temperature, circulation and climate.*

Authors' Reply to 3.: The purpose of this paper was only to focus on the direct atmospheric composition changes caused by GCRs. The effects of GCRs on temperature and circulation had already been discussed in the Calisto et al. (2011) and Semeniuk et al. (2011) papers. In those papers, it was clear that the GCR-caused temperature and circulation modifications also had an influence on the compositional changes. We wanted to cleanly study the GCR-caused compositional changes only, thus it was outside the scope of this paper to also study the GCR impact on temperature and circulation.


Referee #2 - *4. Analyzing the results of sensitivity studies with their 2-D model the authors consider only global/annual mean total column ozone (GAMTCO). I think it is not a good choice because in the tropical area which contributes a lot to global mean value the influence of GCR is very small due to high cutoff rigidity. Therefore the magnitude of the GAMTCO changes caused by GCR is very small. It can be even considered negligible, because it is smaller than the measurement uncertainties. Would*

it be the same if the authors look at the higher latitude zones where the ionization by GCR is more pronounced.

Authors' Reply to 4.: We computed the GCR impact on the annual average global total ozone (AAGTO) because GCRs impact the atmosphere at all latitudes. We agree that the largest impact of the GCRs is at the highest latitudes. We have computed the GCR impact at polar latitudes only (60-90 degrees South and 60-90 degrees North) and present them in the new Figure 9. There are many similarities in shape between the annual average polar total ozone (AAPTO) and the AAGTO, however, the AAPTO is always larger. For example, from the bottom plots of Figure 8 (old Figure 7) and new Figure 9: In 1960 the AAGTO is computed to be -0.13% while the AAPTO is computed to be -0.18%. In 2010 the AAGTO is computed to be -0.11% while the AAPTO is computed to be -0.27%. Thus, the polar differences tend to be larger by the end than they were at the start of the simulation period.

Authors' modification of Paper as a result of 4.: This is now discussed in Section 5.2 in the new fourth paragraph, which reads:

The GCR-caused atmospheric changes are larger at higher latitudes, thus we also compute the annual average polar total ozone (AAPTO). The AAPTO is calculated using the model output only at polar latitudes (60-90 degrees South and 60-90 degrees North) and is given in Figure 9. Both the AAGTO (Figure 8) and the AAPTO (Figure 9) have similar shapes for the total ozone change in the two regions plotted (1000 to 100 hPa and 100 to 1 hPa). In 1960 the AAGTO for the entire troposphere and stratosphere (1000 to 1 hPa) is computed to be -0.13% (see Figure 8, bottom) while the AAPTO is computed to be -0.18% (see Figure 9, bottom). In 2010 the AAGTO for the troposphere and stratosphere is computed to be -0.11% (see Figure 8, bottom) while the AAPTO is computed to be -0.27% (see Figure 9, bottom). Thus, the polar differences tend to be larger by the end than they were at the start of the simulation period.
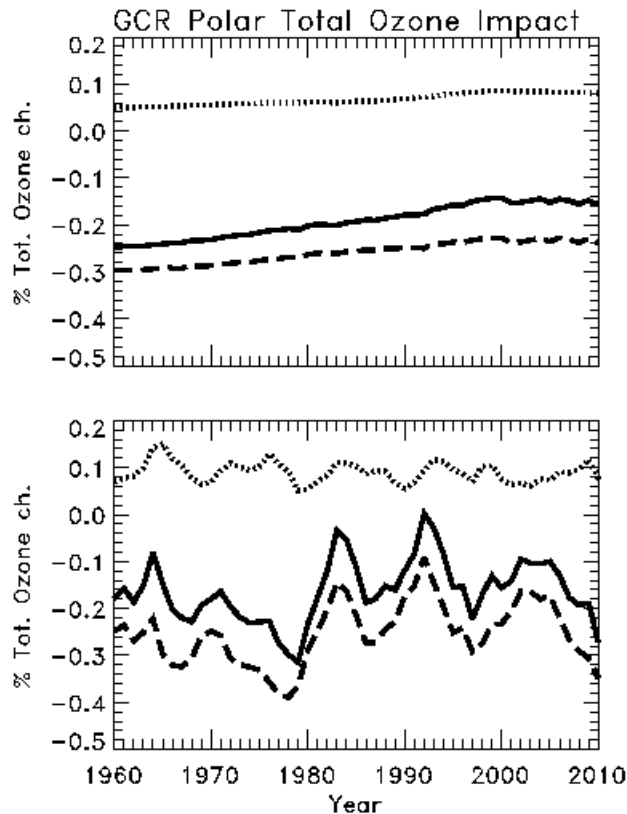
New Figure 9 caption:

GSFC 2-D model GCR-computed impacts of annual average polar total ozone (AAPTO) between 1000 and 100 hPa (dotted black), between 100 and 1 hPa (dashed black), and for the entire troposphere and stratosphere, 1000 to 1 hPa, (solid black) over the 1960-2010 time period. The top plot shows the comparison of simulation A1_GCR_GSFC to A_Base_GSFC. The bottom plot shows the comparison of simulation E_GCR_GSFC to E_Base_GSFC.

Referee #2 Minor/technical issues:

Referee #2 - 1. page 33935, line 26: It reads like GCR produce constituents w/o ionization. I suggest reformulate, because NOx, HOx production is the results of ionization.

Authors' Reply to 1.: The GCRs can produce NOx without ionization. Charged particles can directly dissociate molecular nitrogen (N2) into nitrogen atoms. For example, the N atoms, especially in excited states N(2D) or N(2P), can react quickly with molecular oxygen (O2) to form NO + O. N+ atoms can also lead to production of NO+. These processes are discussed in G. Brasseur and S. Solomon, Aeronomy of the Middle Atmosphere, D. Reidel Publishing Company, 1995, especially see Chapter 6. A more detailed discussion of the dissociation of N2 and O2 by very energetic protons and the associated secondary electrons is given in H. S. Porter. C. H. Jackman, and A. E. S.

Green, Efficiencies for production of atomic nitrogen and oxygen by relativistic proton impact in air, The Journal of Chemical Physics, 65, 154-167, (1976).

It is true that HOx production requires complex chemistry involving positive ions. This is explained in Section 3.

Since NOx can be produced through direct dissociation of N2 (and without ionization), we have not changed the Section 3 title.

Referee #2 - *2. page 33937, lines 5-19: Are lightning and aircraft emissions included in WACCM?*
The authors said they are included in 2-D GSFC.

Authors' Reply to 2.: Yes, lightning and aircraft emissions are included in WACCM (see section 3.4 of Lamarque et al.,The CAM-chem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, Geosci. Model Dev., 5, 369–411, 2012). This is now noted in Section 4.1.

Authors' modification of Paper as a result of 2.: This is now discussed in the revised manuscript at the end of the second paragraph of Section 4.1, which reads:

Tropospheric NOx production from lightning and aircraft is included as described in Lamarque et al. (2012).

Add Lamarque et al. (2015) to the Reference list:

Lamarque, J.-F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. H., Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.,The CAM-chem: description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, Geosci. Model Dev., 5, 369–411, doi:10.5194/gmd-5-369-2012, 2012.

Referee #2 - *3. Section 4.2: How good is representation of tropospheric chemistry in 2-D environment? The chemistry is non linear, but it is necessary to use zonal mean fields. It would be interesting to compare OH distribution from the two applied models.*

Authors' Reply to 3.: The speed of the 2-D model makes it a valuable tool in this study, which includes a number of multi-decadal simulations. We agree that a 2-D model may not represent tropospheric chemistry as well as a 3-D model. However, the GSFC 2-D model's troposphere has been improved recently as was described in section 4.2 (pp. 33938-33939). Since the reviewer specifically asks about OH, we also note that for the current paper, the model tropospheric OH is specified from the monthly varying OH field documented in Spivakovsky et al. (2000). The GSFC 2-D model recently participated in a SPARC Project investigating the "Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species" edited by M.K. W. Ko et al. and published in December 2013 (SPARC Report No. 6, WCRP-15/2013). This report is available online at http://www.sparc-climate.org/publications/sparc-reports/sparc-report-no6/. A profile of OH values is given in Figures 5.10 and 5.11 of that report and shows the GSFC 2-D model with reasonable agreement to the five different three-dimensional models with chemistry.

The annual average OH distributions for year 2009 from WACCM and the GSFC 2-D model are given below. Both models show larger tropospheric amounts in the tropics (~0.05 to 0.2 pptv) and lesser amounts at higher latitudes (~0.005 to 0.1 pptv). Generally, the two models' tropospheric OH abundances are within 30% of each other, although the GSFC 2-D models' upper tropospheric OH amounts can be up to 50% less than WACCM values for mid to high latitudes.
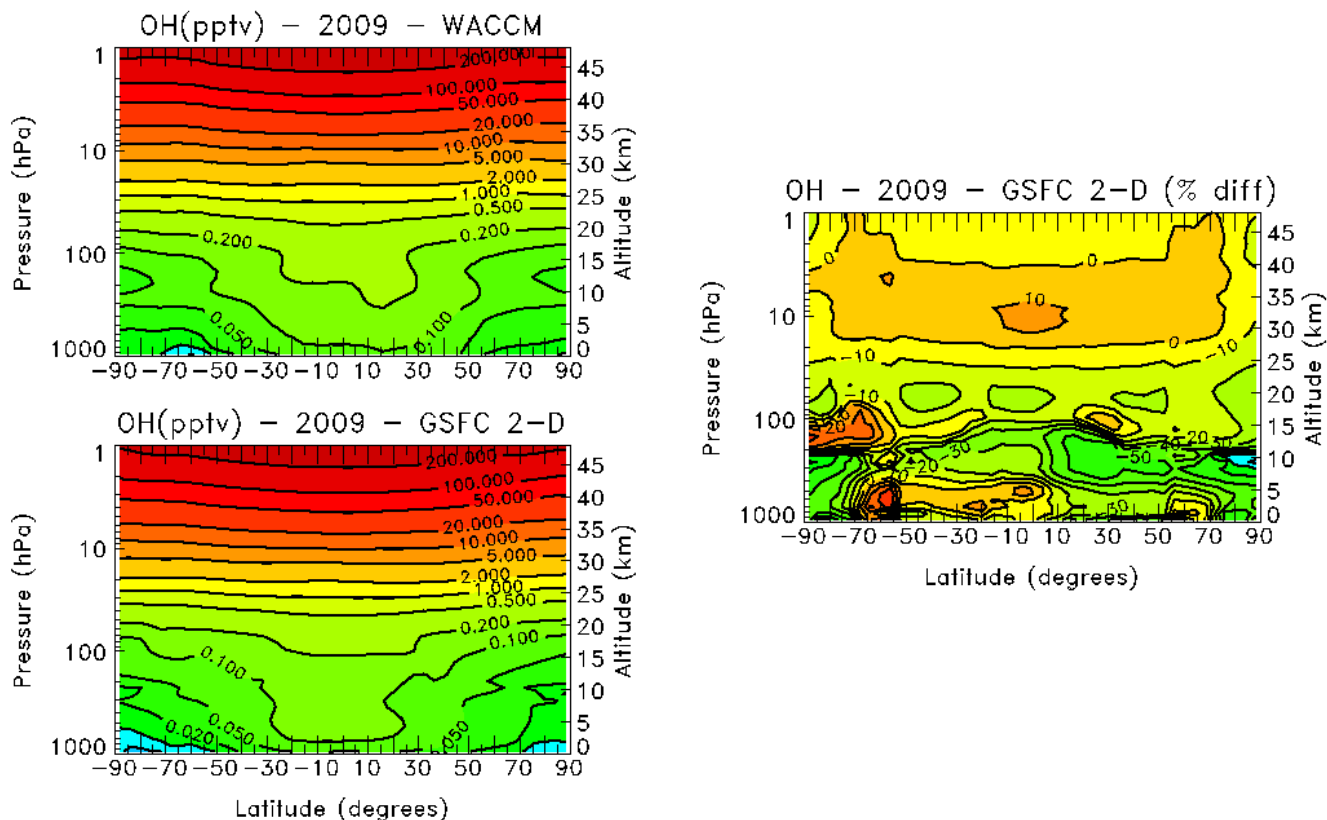
Figure on Comparison of Models' OH.
Annual average OH distributions for year 2009 from WACCM (top left) and the GSFC 2-D model (bottom left) are shown. Percentage difference of the GSFC 2-D model OH from WACCM OH (right) is also shown.

Authors' modification of Paper as a result of 3.: A sentence (third) has been added in paragraph 4 of Section 4.2, which reads:
The model tropospheric OH is specified from the monthly varying OH field documented in Spivakovsky et al. (2000).
Add Spivakovsky et al. (2000) to the Reference list:
Spivakovsky, C. M., Logan, J. A., Montzka, S. A., Balkanski, Y. J., Foreman-Fowler, M., Jones, D. B. A., Horowitz, L. W., Fusco, A. C., Brenninkmeijer, C. A. M., Prather, M. J., Wofsy, S. C., and McElroy, M. B., Three-dimensional climatological distribution of tropospheric OH: Update and evaluation, J. Geophys. Res., 105, 8931-8980, doi:10.1029/1999JD901006, 2000.

***Referee #2** - 4. page 33940, lines 22-23: Not proper explanation. I think NMHC and VOC's included in the both models also play important role.*

      Authors' Reply to 4.: NMHC and VOC's are specified in the GSFC 2-D model using output from a three-dimensional model (the Global Modeling Initiative's (GMI) combined stratosphere-troposphere chemistry and transport model), see the fourth paragraph of Section 4.2. A four-year average (2004-2007), which changes seasonally but repeats yearly, of the GMI model's output is used. Thus the annual average values of NMHC and VOC's do not increase over the 1960-2010 time period.

      We do, however, agree with the referee that our explanation is not totally accurate. We have analyzed this issue by completing several sensitivity studies with the GSFC 2-D model using different temporally changing levels of CH4. We have found that the CH4 increase is only responsible for about one-third of the "tropospheric" ozone increase over the period 1960-2010. In the process of investigating this issue, we realized that the "tropospheric column" line was mislabeled in Figure 7. This line represents ozone at pressures greater than 100 hPa, which is the troposphere in the tropics, but overestimates the tropospheric extent at higher latitudes. We have redone some of the discussion in Section 5.2 to correct this.

      Besides CH4, we found that the increase in chlorine levels over this time period also led to an increase in column ozone between 1000 and 100 hPa. Chlorine thus becomes more important in the control of ozone over the 1960-2010 time period. This means that the formation of ClONO2 through the reaction ClO+NO2+M $\rightarrow$ ClONO2+M becomes more influential when GCRs produce NOx. Thus, the GCRs become more important in affecting ozone variation in the lowest part of the stratosphere (at pressures greater than 100 hPa) at higher latitudes over those 51 years.

      Authors' modification of Paper as a result of 4.: The second sentence of the fourth paragraph of Section 4.2 is modified to read:

      For this, the following quantities are specified using a four-year average (2004-2007) output from recent simulations of the Global Modeling Initiative's (GMI) combined stratosphere-troposphere chemistry and transport model…

      The third sentence in Section 5.1 is modified to read:

      The GCR-caused ozone increase is due to two processes: 1) the GCR-produced NO reacts with CH4 oxidation products (see, also Krivolutsky et al., 2001); and 2) the GCR-produced NO2 reacts with ClO to form ClONO2 and reduces the chlorine-caused ozone loss.

      The first three paragraphs of Section 5.2 are also modified (because of the mislabeling problem noted above) to read:

      The GSFC 2-D model gives fairly similar results to SD-WACCM (compare Figs. 4 and 5) and is significantly faster computationally to use for longer-term simulations. Thus, the GSFC 2-D model was used in several sensitivity study simulations described in Table 1 (and Sect. 4.2) to investigate the longer term GCR-caused changes, particularly focusing on annual average global total ozone (AAGTO) as well as global column ozone in the two regions between 1000 and 100 hPa and between 100 and 1 hPa. The GCR-caused change in ozone in those two regions, separately, and for the entire troposphere and stratosphere (1000 to 1 hPa) is computed for two pairs of scenarios: (1) Fig. 8 (top) shows a comparison of the first pair (A1_GCR_GSFC to

A_Base_GSFC), which are simplified representations of the atmosphere with a climatological mean transport (changes daily, but repeats yearly) in both scenarios and a mean GCR input (constant throughout the simulation) in A1_GCR_GSFC; and (2) Fig. 8 (bottom) shows a comparison of the most comprehensive pair (E_GCR_GSFC to E_Base_GSFC), which include interannually varying transport, sulfate aerosol surface area, and solar cycle photon flux variation in both scenarios and an interannually varying GCR input in E_GCR_GSFC.

First, focus on the results intercomparing scenarios A1_GCR_GSFC to A_Base_GSFC (see Fig. 8, top): the GCR-caused column ozone between 1000 and 100 hPa showed an increase from +0.03% up to ~+0.05% over the 1960–2010 time period, driven partly by increases in CH4 over those 51 years. The GCR-caused column ozone between 100 and 1 hPa also showed a time dependent increase, but started in year 1960 at -0.19% ending up at -0.12% in year 2010. The GCR-caused total AAGTO follows the increases in the two regions noted above, starting at -0.16% in year 1960 and increasing to ~-0.07% in year 2010.

Second, intercompare the more complete simulations E_GCR_GSFC to E_Base_GSFC (see Fig. 8, bottom): the GCR-caused column ozone changes between 1000 and 100 hPa showed a significant variation from ~+0.03% to ~+0.07% over the 1960–2010 time period. The GCR-caused column ozone changes between 100 and 1 hPa also showed substantial variation giving -0.23% in 1979 and -0.02% in 1992. The GCR-caused total AAGTO followed these variations, with a low of -0.19% in 1979 and a high of +0.03% in 1992.

Figure 8 (old Figure 7) caption has been modified (because of the mislabeling problem noted above) to read:

__Figure 8.__ GSFC 2-D model GCR-computed impacts of annual average global total ozone (AAGTO) between 1000 and 100 hPa (dotted black), between 100 and 1 hPa (dashed black), and for the entire troposphere and stratosphere, 1000 to 1 hPa, (solid black) over the 1960–2010 time period. The top plot shows the comparison of simulation A1_GCR_GSFC to A_Base_GSFC. The bottom plot shows the comparison of simulation E_GCR_GSFC to E_Base_GSFC.

__Referee #2__ - 5. page 33943, second paragraph: In Figure 7(upper panel) the increase of tropospheric ozone is explained by CH4 increase. Why it is not the case for Figure 7 (lower panel). It would be interesting to explain.

Authors' Reply to 5.: We have tried to investigate this model predicted variation of "tropospheric" ozone from one year to the next. As indicated above, we plotted ozone between 1000 and 100 hPa with the dashed line rather than "only" tropospheric ozone. Thus, variations in the lowest stratospheric amounts of ozone also have an impact on this variation. As discussed in the paper, the background total chlorine, aerosol surface area, and solar cycle variation of the GCR impact can also have a large influence on the ozone variations. For example, the increase in ozone between 1000 and 100 hPa from 1960 to 1965 is mainly influenced by the increase in GCR NOx production during solar minimum of the mid-1960s and the increase in aerosol surface area in 1963-64. These two processes increase ozone through the following: 1) The

GCR NOx production leads to a tropospheric ozone increase primarily through reaction with CH4 (as explained in Krivolutsky et al. 2001); and 2) the increase in aerosol surface area leads to a decrease in stratospheric ozone loss due to the NOx catalytic cycle, thus any increase in NOx (such as through GCRs) would cause less ozone destruction.

Referee #2 - 6. page 33945, line 21: I think "intensity" should be added after "reactions"

Authors' Reply to 6.: The word "intensity" has now been added after "reactions".
Authors' modification of Paper as a result of 6.: The third sentence in section 5.2.2 has been modified to read:
Enhanced aerosol surface area results in an increase in heterogeneous reactions' intensity on the sulfate aerosols.