

We thank the two anonymous referees for their very useful comments. We have addressed the points raised by the reviewers and updated the manuscript accordingly, please see below for specific responses to each of the points raised.

Reviewer 1

1. Large scale aspects as the position of the descending branch of the Hadley cell in summer over the Mediterranean (J.J. Liu et al., 2009 –JGR) leading a generalized strong subsidence (with ozone enriched air masses from upper levels) with associated free cloud conditions and high radiation, should be mentioned in the introduction.

The reviewer points out an important omission in our discussion of the Mediterranean ozone budget. We have added text to the introduction describing the impacts of ozone imported from the stratosphere and the descent of ozone-rich air from the upper troposphere over the Mediterranean, with reference to studies highlighted by the reviewer (also in response to points 2/3/4 below).

2. A major concern is the total lack of comments and references about the potential role played by natural processes, such as stratosphere-troposphere exchange, in the ozone budget over the Mediterranean region. This prevents proper assessment results on the agreements found between observations (mainly from satellite) and modeling. Specifically, the TOMCAT model considers folding tropopause processes associated to deep lows, cut-off-lows (COL) or to the subtropical jet?

3. Associated with the previous question, Nieto (2005) produced a climatology of cutoff lows over the northern hemisphere from 1958-1998 which showed a favorable area for COLs over three areas of the Northern hemisphere being one of them located over the Eastern Mediterranean Sea where closed cyclones were more prevalent (see Nieto et. al Fig. 8). As you know COLs can ozone enrich the middle and upper troposphere, and consequently the tropospheric column. These results agree others obtained by Gerasopoulos et al. (2006) and Akritidis et al.,(2010) in which they show that the stratospheric intrusions can even affect the O₃ mixing ratios at the ground over the Mediterranean (Northern Greece and Athens, respectively).

4. Weigel et al. (2012; ACP) also documented stratospheric intrusions associated to the subtropical jet (STJ) over the Mediterranean. The STJ can be found over North Africa and the southern edge Mediterranean basin in summer time, and might favor stratospheric ozone-rich air masses transport into the upper troposphere.

We agree that ozone from the stratosphere could be an important source of ozone to the summertime Mediterranean troposphere (see modification of Introduction, also in response to point 1 above). However, as pointed out by Reviewer 2, this source remains the same in all of our simulations. This means that the results of our emission perturbation experiments are valid irrespective of the source of other background ozone in the region, which will likely have some contribution from the stratosphere. Our aim here is to demonstrate sensitivities of Mediterranean ozone to changes in different global and regional sources of ozone precursors. We have clarified that this is the focus of our analysis by modifying the paper title (see Response to Reviewer 2 below). While quantifying the contribution to Mediterranean ozone from the stratosphere is interesting in its own right, it is beyond the aims of our analysis.

5. It would have been appropriate to have included an ozonesonde stations (ie. Thessaloniki) for both model validation and ozone vertical structure assessment since most of results refer to levels well above the marine boundary layer. Why ozonesonde statistics have not been used?

We agree that comparisons to ozonesonde data would have been useful in this study. Unfortunately there are no ozonesonde data over the region for the time period examined in this study. We contacted the PI for the Thessaloniki station suggested by the reviewer and he confirmed that there were no ozonesonde launches from this station for the time period 2005-2008. The only station which falls within the geographic boundaries of our study for which data was available from the WOUDC is Ankara, but with only a handful of launches in this time period the comparison would have little statistical significance.

6. Concerning the anthropogenic emissions you have used IPCC R5 2000 emissions set. Does it take into account the emissions from crude oil refineries, phosphate-based fertilizer industry and power plants settled in North Africa (Morocco, Algeria, Tunisia). See Rodriguez et al (2011, ACP).

We have used the latest IPCC AR5 anthropogenic emissions, in common with many other global modelling groups. These include a comprehensive integration of several regional and global emission datasets (for details see Lamarque et al., 2010 and references therein). Since the emissions data mentioned by the reviewer were published in 2011, they are likely not explicitly included, however we don't believe that their inclusion would affect our conclusion, since we are already showing that local NO_x emissions dominate surface level ozone, and it is meteorology that prevents local emissions from having a strong impact on the mid and upper troposphere, irrespective of the magnitude of the local emission sources

7. Concerning the TOMCAT model experiments, you have eight sensitivity runs, each with 20% reductions in different local and global ozone precursor emissions detailed in Table 2. My first question is, what are the criteria to choose 20% reduction and not 15% or 25%?. It should be explained. My second question: a higher reduction, for example of 30% in anthropogenic VOCs, would be result in major/significant impact on ozone?

We investigate sensitivities to 20% emission reductions, since these have typically been employed in past studies (including the UNECE HTAP (Hemispheric Transport of Air Pollutants) report and resulting publications), and are of a magnitude that may be considered reasonable in terms of achievable emission reductions in response to legislation. In addition, a recent study (Wlid et al., 2012) showed that over the range +/- 60% NO_x emission changes, the impacts on ozone in local and downstream continents are linearly scalable. We therefore expect our results to allow an estimate of the response of Mediterranean ozone across a range of emissions change magnitudes. We have added text to Section 4 as a brief justification of this.

8. Concerning the sensitivity runs, the Asian emissions constitute a very interesting and novel bet. However, I wonder you have not considered the North America emissions

taking into consideration the high number of publications dedicated to the impact of ozone and precursors transport from North America into the Mediterranean. You have referenced the importance of ozone and precursors abundances enhancement in upper and middle troposphere over the western Mediterranean by large-scale westerlies in summer (Rodwell and Hoskins, 1996,2001).

Again, the reviewer makes a good point. As we described in our introduction, North American emissions have been shown to contribute to tropospheric ozone in the Mediterranean free troposphere. For completeness, we have performed an additional simulation to examine ozone response to a North American emissions perturbation. We compare the impacts of this perturbation to the others in terms of the ozone change and associated radiative impacts (see Figures 6 and 8, and section 4.2).

9. In Section 4.2 (Global contributions) you give impact of ozone in pressure levels that are close to the tropopause or well above it. According to NCEP reanalysis, the tropopause is found at pressure levels higher than 150hPa in the north Mediterranean (computed for Jun-August of 2005-2008 period).For example you estimate an ozone impact from anthropogenic NO_x of 1.94 ppbv at around 150hPa in the north and east. In case of Asian emissions, ozone changes of up to 6.09 ppbv are shown for an altitude of 130hPa. In case of anthropogenic emissions (page 27231, line 20) you give ozone change increases of 0.69 ppbv at 100 hPa, and in case of biogenic VOCs, you report an increase of 3.44 ppbv at 100 hPa, as well, levels clearly in the stratosphere. Does it make sense?, do you mean that this impact is observed in the lower stratosphere?. Do you account for troposphere-to-stratosphere transport? These results should be reviewed because, although the eastern Mediterranean, further south, shows in summer a high tropopause (with pressure less than 130hPa, typical from subtropical latitudes), the rest of the Mediterranean (Central and Western) show a mean-latitude tropopause with mean pressure levels above 150hPa. In case you refer to the lower stratosphere a detailed explanation should be provided.

We have calculated the position of the tropopause for the summertime Mediterranean region based on the ECMWF potential temperature (< 380 K) and potential vorticity (< 2 PVU) used to drive the TOMCAT simulations. The Mediterranean region tropopause pressures range between 70 and 130 hPa, depending on month and latitude. We have then used these tropopause pressures to “white-out” the stratosphere in the zonal and meridional average plots presented in Figures 5, 6 &7. This ensures that we are only presenting ozone changes from the troposphere. In addition, we have ensured that our discussion in the main text is limited to the impacts of the emission perturbations on ozone at pressures within the troposphere.

Minor comments/corrections:

Page 27221, line 27: : : : ozone measured in Crete, Eastern Mediterranean: : : should says: : : : ozone measured in Crete (Eastern Mediterranean): : :

The text has been updated.

Page 27222, Line 9:...suggest that his local: : : should says: suggest that this local: : :

The text has been updated.

Page 27223 and the rest of the text, I would replace “Sect.” by “Section”.

Our original manuscript used the word 'Section', it was changed to 'Sect.' by ACPD typesetting.

Page 27228, Lines 16-18: It seems the bias or the SD is missing. Please, revise the sentence is quite confusing.

The sentence has been changed to make it clearer.

Page 27234, Line 3: "A monthly average: : :" should say "A monthly average: : :"

The text has been updated.

Page 27247, Table 2 caption: Replace "mWm-3" by "mWm-2".

The text has been updated.

Page 27251, Figure 5: Latitudinal ozone cross section doesn't fit the latitude range marked by the white box.

The figure has been updated so that the white box now matches the zonal mean plot.

Page 27252, Figure 6: The same as before.

The figure has been updated so that the white box now matches the meridional mean plot.

Page 27254, Figure 8: Remove decimals in the color scale

The decimals have been removed

Font size should be increased in axis and labels of Figures 1, 2, 3, 5, 6 and 7, and especially in lower panels of Figures 2 and 3. It is really very hard to read numbers!

The font size has been increased in all figures.

Reviewer 2

My only strong comment is on the lack (and need) to attempt to explain/discuss potential mechanisms and/or pathways that bring about the main results of the emission contributions. For example, what could be the reason of the prevailing effect on ozone of NO_x among the various emissions? Or, why the global biogenic VOC effect is larger above 700 hPa?

We thank the reviewer for pointing out this omission. Text has been added to sections 4.1 and 4.2 to discuss the mechanisms and pathways which lead to the modelled ozone changes.

For the meteorological contributions I would not worry so much because their effect (of whatever magnitude, see next sentence) is the same in the control and the reduction simulations. However, the meteorology role could be important (as the other reviewer points out) and, since this is not diagnosed in this study, I suggest to slightly modify the Title in order to clarify that only chemical emission sources are examined in this work.

We agree that the title may have been confusing and it has been changed to ‘The Mediterranean summertime ozone maximum: global emission sensitivities and radiative impacts’ so as to more closely represent the study.

Specific Comments

page 27222, lines 1-10: the description of the western Mediterranean ozone is qualitative, limited to the topographical causes of its formation and, in contrast to the eastern Mediterranean in the previous page, does not provide any quantitative information on the observed ozone levels. Please restore this imbalance if possible, by extracting actual values of tropospheric mix. ratio or partial column of summer ozone for western Mediterranean from the already cited papers (e.g. Velchev et al., 2011) or/and others (e.g. Ravetta et al., 2007).

We agree with the Reviewer’s point. We have modified the Introduction to include a more quantitative description of summertime ozone in the Western Mediterranean.

page 27224, TOMCAT description: Please state if the model chemistry interacts with radiation and dynamics.

TOMCAT is an offline chemical transport model, and does not calculate its own dynamics and radiation. It reads meteorology data from archived meteorological service analyses (here from ECMWF). Therefore, there is no interaction between the model chemistry and the model dynamics.

page 27224, TOMCAT description: is chemistry (e.g. in-plume destruction of NO_x) from shipping emissions treated specially (like, for example, in Duncan et al. (2008)?

This type of plume chemistry parameterization is not considered explicitly. Although, we do include emissions from shipping.

page 27226, Evaluation of TOMCAT using satellite observations: Why comparison with TES is done only for 2005? Can other years be included? The same for the GOME-2. More years could give a better feeling for the degree of the observed interannual

variability and the persistence of the summertime ozone maximum (and model capability to capture this).

In order to save on the number of figures / space, we limited the satellite / model comparisons that we showed to one year each for TES. We agree that it may be useful for the reader to be able to refer to the comparisons for all years. We have therefore added comparisons for TES for the years 2005-2008 inclusive to figure 2 showing each individual year's comparisons. The comparison with GOME-2 (figure 3) has been conducted for only 2 years as 2007 is the first year for which data are available and our model run ends in 2008 as this is the last year for which we have GFED biomass burning emissions.

page 27229, line 25: what's the geographical definition of east and west Mediterranean?

Longitude boundaries have been added to the text.

page 27234, line 3: separate "monthly" from "average" (currently appear as one word).

The text has been updated

page 27247, Figure 1 caption: please insert here too the four station names to assist fast inspection and interpretation.

The caption has been updated the include the station names.