Responses to the comments of Reviewer #3

We would like to thank all three reviewers for their comments and for motivating us to significantly improve our study as outlined below:
The present modeling study evaluates the impacts of Istanbul and Athens anthropogenic emissions on the Eastern Mediterranean air quality, based on recently developed emission inventories, background levels, chemical regimes and transport, as well as quantifying the potential impacts of country-based emission mitigation options on the major pollutant levels in the region. The meteorological conditions during the studied period represent the characteristics of summertime and wintertime climatology in the region. Wintertime simulations have been added in order to provide information on the seasonality of the impacts of emissions in the region. This addition is important since although ozone exceedences in the studied region are of concern mainly in summer, particulate matter exceedences largely occur in winter. To our knowledge there is no other such study for the area.
To enable the evaluation of the model performance in particular for the wintertime simulations, we have changed the simulation year to 2008 when data are available. Meteorological characteristics are similar in the two summer periods (2008 for the revised paper and 2004 for the ACPD paper). However, air circulation patterns and temperatures in winter are different from those in summer.
We also thank the reviewers for attracting our attention to the miscalculations regarding the NMVOC/NOx molar ratios reported in the ACPD paper. We have now corrected these ratios throughout the manuscript. This has largely increased the consistency in the discussion.
Finally, we have improved the mitigation scenarios using that developed by the IIASA group for the CityZen project. The new Figure 1 shows the extended areas and the model grids characterized as urban and rural for the discussion of the results.
The results are now presented in a standalone paper with thorough discussion supported by additional material in the supplement. To accommodate the wintertime simulations, the word ‘summertime’ has been removed from the title of the paper.
Our results point out that mitigation can be efficient for pollutants that are mostly of primary origin like CO and $PM_{2.5}$ but the system is more complex for secondary pollutants and in particular for O_3 due to the non linear behavior of the chemistry of O_3-NOx-NMVOC system as documented in the discussion.

We provide here below point-by-point replies to the reviewer comments.

Comments

Certainly, the relationship between air quality and emissions at regional scale is a topic of great interest for the scientific community and environmental stakeholders that deserves a strict scientific analysis. Unfortunately, according to this reviewer this study does not provide relevant information about this issue. A detailed analysis based on field observations and modeling simulations is needed to understand such complex phenomena. The manuscript needs a considerable revision to provide useful information to the community. The authors need to evaluate the convenience of publishing such particular application of a modeling approach that has been already published in previous articles. The manuscript makes reference to another five publications based on the same modeling system and data for the same region. The *current manuscript could be more suitable for a local or internal publication than for a journal such as ACP.*

Response: As outlined in the general statement in the beginning of our replies to the reviewer comments, we largely improved this study in order to address all concerns of the reviewers by performing both winter and summer simulations over 'typical' winter and summer months in 2008 (not summer 2004 as in the ACPD paper and in the two earlier studies). In 2008 observational data are available in the area for model evaluation. We have also used more realistic mitigation scenarios developed within the CityZen project and added further discussion of the results. To our knowledge this is the first modeling study that evaluates the impacts of Istanbul and Athens anthropogenic emissions on the Eastern Mediterranean air quality, based on recently developed emission inventories, background levels, chemical regimes and transport, as well as quantifying the potential impacts of country-based emission mitigation options on the major pollutant levels in the region.

Some major comments are:
- To provide useful information any modeling study needs to evaluate its results with field observations. The manuscript does not present any modeling evaluation. Modeling uncertainties may be higher than the reported differences between scenarios.

Response: We have now added in a new section 3.1 detailed model evaluations for the winter and summer simulations including aerosol chemical compositions. We have changed the simulation year from 2004 to 2008 in order to have more complete observation data to evaluate our model results, particularly for winter since year 2004 is lacking observations in most areas of interest. The observational dataset is described in section 2.3 supported by Table S3 in the supplement that provides information on the geographic location of the observational sites and the pollutants for which observations are available.

- The section describing the methodology and input data is very brief. The authors refer to the reader to at least seven additional articles for basic information about the modeling system and emissions data, which is not practical.

Response: Section 2 on 'Materials and Methods' has been extended to provide all important information for the present study on the model configurations, chemical mechanism for gases and aerosols, emissions, scenarios adopted including mitigation scenarios as well as the observations used for the model evaluation. This section followed by the model evaluation in the new section 3.1 make the paper stand-alone. In addition, changing the year of simulations from 2004 to 2008 to enable evaluation during winter time also, the evaluation of the modeling system is new compared to our earlier published studies.

- A description of the meteorological conditions during the simulated period is needed. Is the selected period representative of typical meteorological conditions during summertime?

Response: We have now added winter time simulations and extended the simulation periods to cover the whole months of interest. We have also provided information on the meteorological characteristics of the region in winter and summer conditions and provided references in order to show that these characteristics are representative of the general patterns in the region in section 2.2. See also reply to comment 1 of reviewer #1 and the comment below.

- *More information about the emissions used as input data is needed. Which methodology was used for the chemical characterization of the emissions (e.g. VOCs speciation)?*

Response: The description of anthropogenic emissions used in the model has been complemented (Section 2.1) also providing details on the NMVOC and PM emissions chemical speciation. Table S1 in the supplementary material shows the winter and summer sectoral distributions to the anthropogenic emissions of CO, NO_x, SO_2, NMVOC and PM_{10} in the Greater Istanbul (GIA) and Greater Athens (GAA) Areas. In addition, Table 1 provides the absolute emissions from GIA, GAA and the entire domain for the winter (December 2008) and the summer (July 2008) months of the simulation, distinguishing between anthropogenic and biogenic emissions. Table S2 in the supplementary material provides country-based emission mitigation factors applied for the mitigation scenario.

Section 2.1 now reads:
'The anthropogenic emissions for the model domain are compiled by merging emissions from Greece on 10 km, Athens on 2km (Markakis et al., 2010a and b) and from Istanbul on 2 km resolution (Markakis et al., 2012) into the emission inventory of the French National Institute of Industrial Environment and Risks (INERIS: https://wiki.met.no/cityzen/page2/emissions), which is a re-gridded product of the 50 km × 50 km European Monitoring and Evaluation Programme (EMEP) inventory (http://www.ceip.at/). The non-methane volatile organic compound (NMVOC) emissions are speciated into 23 species using profiles from the EDGAR global inventory (Olivier et al., 2001) and the particulate matter (PM) emissions are speciated in to organic and elemental carbon, sulfates, nitrates and other species based on CARB (2007) profiles. The NMVOC emissions are then converted to CB05 species using the factors provided by Yardwood et al. (2005). The vertical distribution of emissions is calculated based on the Selected Nomenclature for Air Pollution (SNAP) codes provided by Simpson et al. (2003).
The anthropogenic and biogenic emissions from the Greater Istanbul Area (GIA), the Greater Athens Area (GAA) and the entire model domain (areas are shown in Figure 1) integrated over the winter (December 2008) and summer (July 2008) simulation periods are presented in Figure 2. Details are given in Table 1. GAA has larger NMVOC and CO emissions compared to GIA. On the opposite, GIA has 3 to 4 times higher NO_x emissions, 2 times higher SO_2 emissions and ~2 times higher PM_{10} and $PM_{2.5}$ emissions compared to GAA. Figure 2 also shows that biogenic NMVOCs are ~30 times higher in summer than in winter. However, even during summer, the anthropogenic NMVOC emissions in GIA and GAA are by a factor of 2 and 3, respectively, larger than the biogenic NMVOCs. The molar NMVOC/NO_x ratios in the emissions (Table 1) also suggest a larger potential of O_3 production over GAA compared to GIA in both seasons. Table 1 also shows that GIA and GAA emissions can contribute up to more than 20% of the total anthropogenic emissions, depending on the pollutant.

Table S1 in the supplementary material shows the contribution of major sectors (in %) to the total anthropogenic emissions of the major primary air pollutants and how this changes between winter and summer. These changes are marked by the contribution of heating during winter. It can be seen that in both seasons, on-road traffic is a major source of air pollutants, particularly for CO and NO_x. Industry (including energy) is important for SO_2 and PM. Solvent use and traffic are the major contributors to NMVOC emissions. Table S1 also shows the importance of maritime emissions, particularly in Athens.'

- *Which photochemical mechanism was used?*

Response: The gas phase chemical mechanism used for the present study is Carbon Bond- V (CB05) (Yardwood et al., 2005), the AERO5 aerosols modules and aqueous chemistry (Foley et al., 2010) have been employed in CMAQ. In this model configuration, anthropogenic secondary organic aerosol (SOA) is produced by oxidation of toluene, xylene and benzene while biogenic SOA is formed through oxidation of monoterpenes, sesquiterpenes and isoprene. There is also aqueous-phase production of SOA through glyoxal. CMAQ assumes no volatility of primary organic aerosol, in its present configuration does not account for SOA formation from intermediate volatility organics and treats all SOA species as semi-volatile. More details are provided in Carlton et al. (2010).

We have added this information at the end of the introduction of section 2.

- *Which is the influence of other large urban areas (e.g. Thessaloniki) in the modeled region?*

Response: The present study focuses on the impacts of Istanbul and Athens which are concentrated emission spots in terms of their populations. We do not discuss other large cities individually but we compare the impact of these two hot spots of pollution to the impact of the entire anthropogenic emissions in the model domain. The simulation 'No Anth' that omits the anthropogenic emissions in the entire model domain is enabling the evaluation of the contribution of Istanbul and Athens anthropogenic emissions to the changes due to the total anthropogenic emissions in the region. These results are provided in Tables 5 and 6.

- *Better assumptions are needed for the scenario simulating a hypothetical decentralization of both cities. For example, cities extend following certain urbanization and economic patterns, and not arbitrarily. As cities grow or extend, changes in emissions are expected (e.g. transportation), and therefore the actual emissions cannot be just "re-distributed" in a larger area.*

Response: To address the concerns of the reviewers regarding the mitigation scenario by emissions re-distribution, we have now included a more realistic scenario that simulates the impact of country-based anthropogenic emissions mitigation provided by IIASA. This is now applied to both to summer and winter cases.

The description of this scenario is provided at the end of section 2.2:
g) 'Scenario Mitig: This investigates the impact of emission mitigation following a country-based scenario developed in frame of the CityZen project. It is based on the

energy projections developed within the Global Energy Assessment (GEA) by using the IIASA MESSAGE (Model for Energy Supply Strategy Alternatives and their Global Environmental Impacts) model (Messner and Struberger, 1995). Within the CityZen project, IIASA provided ratios of anthropogenic emissions in LowGWP climate-friendly scenario for the year 2030 to those for present day. LowGWP mitigates the impacts of several pollutants emitted from anthropogenic sources on climate, public health and the wider environment. For this purpose, nearly 2000 measures that consist the GAINS database have been evaluated considering different degrees of reductions of specific pollutant species (e.g. CH_4, CO, EC, OC, SO_2, NO_x, VOC and CO_2). The country-based ratios used in the present study for the countries that are within the model domain are provided in Table S2 in the supplementary material. These ratios are multiplied with the base case anthropogenic emissions for each country and pollutant. For primary aerosol components, factors are provided for $PM_{2.5}$, OC and EC and the $PM_{2.5}$ factors are also applied to the other primary aerosols components. The highest reductions are projected for OC and EC emissions. A reduction of 15-40% in $PM_{2.5}$ emissions is calculated while the reduction is up to 10% for SO_2, 3-30% for NO_x, 8-23% for VOCs and 5-40% for CO.

The discussion on the impact of mitigation on air quality is appropriately modified in section 3.5 and supported by Figures 8 and 9 and Table S5 in the supplementary material. The main conclusions of our study remain unchanged, i.e. that mitigation can be efficient for pollutants that are mostly of primary origin like CO and PM2.5 but the system is more complex for secondary pollutants and in particular for O_3 due to the non linear behavior of the chemistry of O_3-NOx-NMVOC system as earlier discussed and now better documented in our revised manuscript.

- The use of many acronyms and percentages to describe the modeling results makes difficult the manuscript reading. Some editorial mistakes complicate also the results understanding, for example: Page 26667, lines 20-21. There is a big difference between 2.92 ppb and 25 ppb. Page 26674, line 19. A maximum contribution of 100% could be expected, but not a contribution of 229%.

Response: We have rephrased large parts of the discussion section for clarity and in order to increase its readability. All needed acronyms are now explained properly.