Dear anonymous referee #1,

Thanks a lot for considering our manuscript suitable for publication after revision and also for the comments and suggestion that you gave us, especially those on the stratospheric O_3, which improved a lot the article. As you will see in the revised version we took into account all your valuable comments and suggestions. We also attach below a report on changes addressed and reply to your specific queries.

Following one of your suggestions, to evaluate the stratospheric episode, we requested the support of Prof. M. Millán, who accepted and contributed a lot; so we have included him as a co-author in this revised version.

Thanks a lot for your constructive review.

Xavier Querol on behalf of all co-authors

Report on changes addressed and reply to specific queries

Overview: The paper deals with the phenomenology of the summer ozone episodes over the greater area of Madrid, Spain. I think that it is a very interesting study, analysing atmospheric measurements of ozone and fine particles together with many other atmospheric parameters and giving further insight to the complicated atmospheric mechanisms related with air pollution over the area. In my opinion, the document deserves publication in ACP, after the recommendations listed below are taken into account. REPLY, Thanks a lot for considering our manuscript suitable for publication after revision and also for the comments and suggestion on the stratospheric O_3 that improved a lot the article.

General comments: A weak point of the paper is that the levels of measured surface ozone are mainly related (or attributed) to the photochemical ozone production over the metropolitan area of Madrid. REPLY, Thanks a lot this comment. We also considered long range transport (named 'external' in the paper. But as you might see in the revised version we have enlarged this and also the stratospheric issue that we miss in our original submission.

On the other hand, I think that the variations of the background ozone levels within the boundary layer and the free troposphere are not discussed with sufficient detail. In relation to that comment and based on research results carried out on the other side of the Mediterranean basin, in the Eastern Mediterranean, it comes out that the regional background ozone levels in the free troposphere and the boundary layer during summer, regularly exceeding 60 ppb, contribute on the average to the greatest part of the surface ozone levels measured in large urban areas like Athens (Kalabokas et al., 2000; Kourtidis et al., 2002; Kouvarakis et al., 2002; Lelieveld et al., 2002; Kalabokas and Repapis, 2004; Gerasopoulos et al., 2005). The main origin of these high ozone background levels over the Eastern Mediterranean is tropospheric ozone subsidence, which seems to be strongly related with specific synoptic meteorological conditions, occurring very frequently during summer at the Eastern side of the Mediterranean basin (Kalabokas et al., 2013; Zanis et al., 2014; Kalabokas et al., 2015; Akritidis et al., 2016). In addition, recent research shows that during springtime ozone episodes (April – May) over the western Mediterranean similar synoptic meteorological patterns might also occur and which are linked with regional episodes mainly induced by large scale tropospheric ozone subsidence, influencing (or fumigating) the boundary layer as well as the ground surface ozone concentrations (Kalabokas et al., 2017).
REPLY, Thanks a lot for these observations. Yes you are completely right; we miss these issues in our discussions and interpretations. We have added these observations and discussions in several part of the revised paper. For example text added or changed includes:

- Abstract: We added: The results demonstrate the concatenation of venting and accumulation episodes, with relative O_3 lows (venting) and peaks (accumulation) in surface levels. Regardless of the episode type, fumigation of high altitude O_3 <u>(from different origins</u>) contributes the major proportion of surface O_3 concentrations.
- Section 1: New text added: In the Eastern Mediterranean, the regional background O3 levels in the free troposphere and the boundary layer during summer might regularly exceed 60 ppb, and fumigation of these upper air masses contribute on the average to the greatest part of the surface O3 levels measured in Greece (Kalabokas et al., 2000; Kourtidis et al., 2002; Kouvarakis et al., 2002; Lelieveld et al., 2002; Kalabokas and Repapis, 2004; Gerasopoulos et al., 2005). Furthermore, a number of studies reported contributions of stratosphere to the surface O3 concentrations during specific

meteorological scenarios in the same region (Kalabokas et al., 2013, 2015; Zanis et al., 2014; Parrish et al., 2012; Lefohn et al. , 2012; Akritidis et al., 2016, among others). In addition, recent research shows that during springtime O3 episodes (April – May) over the WMB similar synoptic meteorological patterns might also occur; and that these are linked with regional episodes mainly induced by large scale tropospheric O3 subsidence, influencing the boundary layer as well as the ground surface O3 concentrations (Kalabokas et al., 2017). However, the most intense episodes in the WMB occur in June-July according to the statistics for the 2000-2015 period by Querol et al. (2016) for Spain.

- Section 3.1. New text added: The AEMET free-sounding shows low O3 surface concentrations (<45 ppb) and high levels (>70 ppb) in the middle troposphere (3000-5000 m a.s.l.), associated with very low relative humidity and intense W to NW winds blowing at that height, which will be discussed in section 4.
- Section 4: discussion added, see below.
- New figure added (see below)
- Conclusions modified (see below)

Even if the typical meteorological conditions prevailing over the Iberian Peninsula during summer are quite different than in the Eastern Mediterranean, as it is very well described in the introduction of the manuscript, occasionally such conditions might occur. In fact, I think that this is the case of the ozone episode of 11-15 July 2016, which is the most studied period in the manuscript (when the intensive measuring campaign has taken place). As shown in Figs 3 and 12, the free tropospheric ozone levels are much higher on July 13, 2016 than the two weeks before and after and at the same time the relative humidity values in the lower troposphere are close to zero (and being in sharp contrast with the periods before and after). In fact, this feature is a very common characteristic of deep and large-scale tropospheric subsidence in summertime ozone vertical profiles over the Eastern Mediterranean, indicating an origin of air masses from the upper tropospheric or stratospheric layers (Kalabokas et al., 2013; Kalabokas et al., 2015).

Therefore, for a better assessment of the free tropospheric influence as well as the reported fumigation events over the area, I would suggest putting more emphasis on the analysis of the synoptic conditions during this most studied period, when the intensive measurement campaign has taken place (11-15 July 2016). A figure could be added including at least the daily meteorological maps of geopotential height, omega vertical velocity and specific humidity at 700hPa pressure level (representative for the free troposphere), which I think that they would be sufficient to follow satisfactorily the evolution and the geographical extent of the subsidence phenomenon (the subsiding air mass seems to originate from N-NW Atlantic). If this parameter is taken into account, then I think that the discussion concerning the origin of the fumigation events during 11-16 July 2016 would be more complete (tropospheric ozone subsidence in addition to the local ozone photochemical production associated with valley-breeze recirculation and ozone residual layers, as mentioned many times in the manuscript). So, I would suggest modifying accordingly the respective paragraphs, where sometimes the high ozone values recorded at the top of the boundary layer are not fully explained (e.g.: Page 5, lines 175-187; Page 9, lines 348 – 352, 362-365; Page 12, lines 470 – 473, 483-487; Page 14, lines 570 – 582).

REPLY. Thanks a lot for these very interesting comments. Indeed we have applied all your suggestions and modified the introduction, discussion and conclusions sections to this end. Examples are:

- Section 4. We added the following discussion: Considering the free sounding O3 profiles in Figure 3, high O3 concentrations (>70 ppb) can be observed above the PBL, between 3000 and 5000 m a.s.l., which may be related to larger scale transport of pollutants, previously uplifted to the mid-troposphere or originated after a stratospheric intrusion and a subsequent deep subsidence into the middle troposphere, as it is probably the case based on the ECMWF ERA-Interim reanalysis data. Transport of high O3 air masses in the middle troposphere, as for the 13/07/2016 in Figure 3, was also documented by Plaza et al. (1997) over this area in July 1994, during the final phase of a high O3 period. More recently, Kalabokas et al. (2013, 2015, 2017), Zanis et al. (2014) and Akritidis et al (2016), among others, have shown that similar transport processes of enriched O3 layers at high altitude, can contribute to increase surface O3 concentrations during the summer in the Eastern Mediterranean. This transport has been associated with large scale subsidence within strong northerly winds in the Eastern Mediterranean (Etesian winds), and the affected layers are dryer than average and show negative temperature anomalies. Figure S11 shows the ECMWF ERA-Interim reanalysis together with the AEMET O3 free soundings at Madrid airport for the 13/07/2016. The ridging at the lower troposphere over the Bay of Biscay at the rear of an upper-level trough (left panels) is accompanied by intense NW winds blowing at the middle and upper troposphere and NE winds at ground level and up to 2000 m (see the radiosonde profile in the same figure). The O3 intrusion is associated to the upper-level trough (Sections A-A and B-B in the figure), and a large area of deep subsidence and extremely low relative humidity observed within the NW flows over Madrid and to the north of the Iberian Peninsula and the Bay of Biscay. High O3 concentrations values and low relative humidity of the ERA-Interim profiles over the airport of Madrid (green and red dotted-lines in the panel "g" of the Figure S11) are in agreement with the radiosonde observations in the same panel.

The question now is how much of this O3 could fumigate at ground level. According to the radiosonde data, the mixing height top was about 2000 m a.s.l. at midday, but could increase to about 3100 m a.s.l. after the projection of the surface temperature increase observed during the afternoon at near-by stations. This height reaches the lower part of the O3 enriched layer originated in the tropopause folding. Thus, a certain impact seems likely. However, the O3 concentrations were relatively low at all surface stations during that day, as it corresponds to a vented low O3 period.

- A new figure S11 has been added and discussed to show the stratospheric intrusion following your suggestion.
- We added these two paragraphs in the conclusions:

- The O3 source apportionment is very complex, having contributions from local/regional and remote sources, including the stratosphere. The relative contributions of these might vary in time and space (e.g. Lefohn et al., 2014).

- Climate change my reduce the benefits of the O_3 abatement policy (since hot waves increase O_3 episodes), and this, as well as the measures and policies in N America and Asia will need to be considered into future Europe policies for O_3 mitigation (Lefohn and Cooper, 2015; Sicard et al., 2017).

- And we modified the fourth as follows: · ·In the MAB, during the highest O3 (accumulation) episodes, in addition of the contribution (to surface concentrations) by fumigation of upper O3 (from regional transport, hemispheric free troposphere O3, and intruded stratospheric O3, X in Figure 10), there is an added fraction produced locally and transported-recirculated within the MAB, which accumulates from one day to the next (Y in Figure 10).

In addition, I think that it would be more appropriate to refer to "Western Mediterranean" ozone when analyzing ozone in Spain (instead of "Mediterranean" or "S. Europe" in general) as, according to the above mentioned papers, the phenomenology of the summertime ozone over the Eastern Mediterranean seems to be is quite different than the typical ozone phenomenology over the Western Mediterranean.
REPLY: Yes we have revised and changed following your suggestion. Thanks for this.

I would strongly recommend taking these considerations into account, which have been made in the spirit to further improve this good quality manuscript, by modifying the respective paragraphs. After responding to these remarks, I think that the paper is ready for publication.
REPLY: Yes we did implement your very good observations and we thank you a lot for your very valuable review.

Technical comments: Fig. 4: Very condensed and difficult to follow, especially on printed paper. I would suggest splitting into two parts and eventually using gridlines.
REPLY: We made it again with clearer drawings.

END OF THE REPORT