Final Author Comments

The authors would like to thank the Editor and the Reviewers for the helpful and constructive comments.

The replies of the authors in each comment are in bold letter characters.

SCIENCE COMMENTS (optional) (ACPD Editor)

SC.1: The low-ozone regime is characterized by reduced static stability in the lower troposphere. The question then arises whether this reduction of static stability is simply an indicator of the low-pressure (cyclonic) condition, or plays a more active role in promoting, e.g., static instability and greatly enhanced vertical transport from the boundary layer. The paper is somewhat too speculative about these possibilities and does not offer firm proof that vertical transport by instabilities is actually occurring. From my perspective, the null hypothesis should be that static stability is tied to the primary circulation via inversion of Ertel potential vorticity, which is partitioned between vorticity and static stability anomalies. As a tracer of large-scale motion, ozone is then associated with the air mass in which it is embedded, and its behavior follows the quasi-horizontal transport of other tracers, and of isentropic PV itself. Rapid vertical transport is unable to explain the observed ozone behavior. Incidentally, since ozone is carried as a dependent variable in the meteorological analyses, you needn't rely solely on MOZAIC data to address this question.

We agree with this remark that during the low ozone regime in the lower troposphere the horizontal transport, mostly in the form of strong westerly air mass flow transporting oceanic air masses from the Atlantic, is also very important. But in any case, upward vertical transport indeed occurs during low ozone cases, as it could be observed in Table 2: About 16% of the air masses arriving at the 1.5 - 5 km layer originate from lower altitudes (0-850hPa) one day before the measurement.

The manuscript will be modified accordingly taking also into account the remarks with a similar content of Reviewer 2 (Comment 2).

SC.2: Regarding sources of ozone, in addition to the stratosphere there's the heavily polluted air over land in the Northern Hemisphere. In such locations, tropospheric ozone maximizes in the upper AND lower troposphere, with a minimum in mid-troposphere. It would be of interest to know whether the northerly flow that brings high ozone from Europe is bringing stratospheric air down from aloft, or lower tropospheric air sideways from the polluted regions (or both). FLEXPART can address this question easily; see, e.g., Trickl et al 2011 ACP for longer trajectory integrations extending up to 20 days.

The mentioned average pattern of vertical ozone profiles is usually observed at more northern latitudes in the European continent but not in the Eastern Mediterranean, as it is observed in Figs. 1, 2 and 6.

Also, from the examination of the FLEXPART back-trajectories corresponding to high ozone days in the lower troposphere, available for MOZAIC profiles (see Figs 3, 5 and Table 1), it comes out that most of the air masses originate from Central Europe but from high altitudes (above 500 hPa), indicating either upper tropospheric or stratospheric origin (>50% of air masses, 3-days before measurement). It is also observed and mentioned in the manuscript that a small fraction of air masses, under the high ozone regime, originate from the boundary layer (<5%) but from over the Balkans or Eastern Europe, eventually transporting air pollution emissions from these areas. K. Schaefer Received and published: 24 January 2013

Did you determine the boundary layer or mixing layer height from your measurements? I think it would be a further important parameter to study the meteorological factors controlling the lower-troposphere ozone levels.

The authors agree that the question of the coupling between the boundary layer and the free troposphere is a very interesting and complicated one. The indicated last phrase of the paper describes the next step in the analysis of the given data-set, which will be the examination of the conditions associated with highest and lowest ozone in the boundary layer. At the same time the possible interaction between the two layers will be examined (in the present paper "lower troposphere" implies actually the 1.5 - 5km layer only). The mid-day boundary layer in our area, especially over the sea is reported to be below 1 km, which is in agreement to what is observed in the changes of meteorological and chemical parameters of the MOZAIC vertical profiles. So, it was beyond the scope of the paper to deal with the boundary layer height, because the layer of our interest (1.5 - 5km) was well above this level.

K. Kourtidis (Referee) Received and published: 31 January 2013

General comments

The authors examine the meteorological (both local and synoptic) conditions that regulate through horizontal and vertical transport the tropospheric ozone levels over Eastern Mediterranean. The topic is of relevance since ozone levels in the area are above the EU air quality standard much of the time during summer and above the EU phytotoxicity limit almost year-round. The work adds to a number of existing studies and clearly and convincingly clarifies the dynamical aspects of this topic. The paper is well written, presents a thorough analysis and is within the scope of ACP. I recommend publication after consideration of some quite minor comments.

Specific comments p. 2460 lines 15-17, "significantly enhanced": as compared to what? Please clarify.

The meaning of this phrase is that the Mediterranean vertical ozone profiles show enhanced concentrations when compared with the quasi-simultaneously measured Central European profiles.

p. 2461 lines 25-26: Instrumentation for NOy is mentioned, but it is not used in the analysis. The authors should remove this reference to the NOy instrumentation (or, even better, present the NOy results also; NOy would be very useful in offering information about the chemical state of the air masses).

Since there are no NOy measurements in the examined profiles, the reference to NOy instrumentation could be removed from the manuscript.

p. 2462 line 9, "36 profiles over Heraklion [: : :] and Rhodes". 36 profiles over each of the two airports? Please clarify.

There are 36 ozone vertical profiles in total, over both airports of Heraklion and Rhodes.

p. 2462 line 18: the boundary layer is claimed to be 0-1 km in general. I would tend to believe that it would be somewhat higher in the area during summertime (0-2 km?).

We agree that the mid-day boundary layer in the examined area over land might reach or even exceed the 2 km. On the other hand, over the sea and based on previous studies it would be considered to be below 1 km, and something similar is observed in the present study after examining the vertical changes of the chemical and meteorological parameters, especially the temperature gradients. It has to be kept in mind also that the examined atmospheric layer in this study (1.5 - 5 km) it is in principle above the boundary layer and it was selected exactly for this reason (avoiding possible interferences caused by boundary layer processes).

p. 2470 lines 25-26: Some reference on how CAPE and CIN were calculated would be useful.

Based on our analysis the tropospheric stability or instability atmospheric conditions associated with highest or lowest tropospheric ozone concern in principle extended air masses covering large geographical areas. So, the most important information on CAPE and CINS parameters of the selected days could be found in the daily data vertical sounding stations of the Eastern Mediterranean Basin. These data as well as the CAPE and CINS calculations derived from them are provided on the Web by the University of Wyoming, College of Enginnering, Department of Atmospheric Sciences (http://weather.uwyo.edu/upperair/sounding.html). As mentioned in the paper, CAPE and CINS values indicating instability of air masses over the area during low ozone days and stability during high ozone days have been observed. In addition, we calculated CAPE and CINS values based on the MOZAIC data and using the general literature formulas, which in fact verified the same pattern.

Technical corrections (All technical corrections will be inserted in the revised version of the manuscript)

p. 2458 line 18: ozone rich -> ozone-rich

p. 2458 line 26-p. 2459 line 1: ozone at ground surface -> ozone near the ground surface

p. 2460 line 3: long range -> long-range

p. 2465 lines 13 and 21, and other parts of the text as well as in Table 1: >500 hPa-> <500 hPa

p. 2465 line 25: Tel Aviv, Israel -> Tel Aviv (for consistency, since in the rest of the text countries of the cities are not mentioned)

p. 2467 line 18: In Cairo profiles: : : -> In the Cairo profiles: : :

p. 2468 line 16: wind speeds [: : :] are higher -> wind speed [: : :] is higher

p. 2469 lines 19-21: hand the -> hand, the
with low-pressure -> with a low-pressure
Europe leading -> Europe, leading
region diffusing -> region, diffusing
the air pollutants, but also are associated -> the air pollutants. They are also associated
(sentence too big)

p. 2471 line 2 In fact -> These line 5 corresponding days radiosoundings -> corresponding radiosoundings line 6 contrary during : : :: : : . calculated -> contrary, high CIN values have been calculated during the days with highest ozone values

Figures 1, 2, 6, 8, 10: The axis numbering is difficult to read

Anonymous Referee #2 Received and published: 21 March 2013

The authors have conducted an analysis of MOZAIC ozone profiles over the eastern Mediterranean region to understand the meteorological conditions associated with high and low ozone levels in the lower troposphere. There is significant interest in understanding the impact of transport on tracer distributions over the Mediterranean since the region is influenced by transport of pollution from Europe, Asia, and North America. The manuscript is well written and the analysis is straightforward. I recommend publication of the manuscript in ACP after the authors have addressed my concerns, which are discussed below.

General Comments

1) A concern with the manuscript is with the discussion of the impact of the meteorological conditions on boundary layer ozone. I found the explanation of this confusing. On page 2469, lines 1-15, the authors explain that under anticyclonic conditions there is northerly flow on the eastern edge of the anticyclone region which transports high ozone from the polluted areas of Europe into the eastern Mediterranean. And that this transport, combined with trapping of the accumulated pollutants in the boundary layer results in high ozone abundances in the boundary layer. However, boundary layer ozone is not higher under anticyclonic conditions. Comparison of Figures 8 and 10 shows that when ozone is high in the lower free troposphere (associated with anticyclonic conditions) ozone in the boundary layer is typically lower than when ozone in the lower free troposphere is low. The difference is especially pronounced over Cairo (Figure 8). When lower tropospheric ozone is high over Cairo, boundary layer ozone is about 30-40 ppb. In contrast, when lower tropospheric ozone is low, boundary layer ozone is about 60 ppb. Over Tel Aviv (Figure 10) high lower tropospheric ozone is associated with surface level ozone as low as 20 ppb. Is it possible that under anticyclonic conditions, the increased stability in the boundary layer results in lower ozone abundances at the surface due to greater ozone loss through deposition at the surface? I appreciate that the authors plan to conduct a more detailed analysis of the boundary layer ozone in a subsequent paper, but the authors need to clarify the discussion here.

In the present paper, the mentioned Figures 8, 10 refer to the Cairo and Tel-Aviv airports, where apparently different conditions in the boundary layer exist, compared to the free lower troposphere. It is also very important to keep in mind that the classifications of high and low ozone days is based on the 1.5 – 5km layer, in order to clarify the mechanisms controlling the ozone variability just above the boundary layer.

The mentioned text at the beginning of this comment is based mainly on the results of the cited publications, referring to previous research and dealing with the boundary layer ozone over the area, mostly at the Aegean Chanel. The above description refers to the boundary layer conditions at these sites, especially the characteristic for this area strong summer northern flow (Etesian winds).

2) A conclusion of the study is that low free tropospheric ozone is associated with the

"uplifting of boundary layer air, poor in ozone and rich in relative humidity to the lower troposphere." The discussion on page 2471, lines 1-7, of the CAPE over Cairo gives the impression that this uplift is local. However, that cannot be the case since over Cairo (as shown in Figure 8) ozone levels are higher in the boundary layer when ozone is low in the lower free troposphere. Furthermore, the back trajectories suggest that the low ozone air originated in the central Mediterranean 3 days before. Are the authors suggesting that the instability conditions at Cairo are representative of the entire Mediterranean region? The authors need to better explain the influence of this uplift on the lower tropospheric ozone levels. Is it possible that the low ozone and high humidity are due to the uplift of boundary layer air from a more remote location? Could this be ozone poor air that is transported from the eastern Atlantic and uplifted by the topography over northwestern Africa? What would 5-day or 7-day back trajectories show for the origin of the low ozone air?

This is really a good and constructive remark helping the discussion. Indeed, the uplifting might not be only local and might concern larger areas, especially those located upwind of the studied airports. The hypothesis of the transport from the eastern Atlantic could be possible and the examination of the composite maps of the days preceding the measurement indicates towards this direction. For the MOZAIC profiles, the calculation of the FLEXPART back-trajectories is done for 3 days before, as shown in the paper. In any case, the main idea is that this westerly flow is associated with low pressure systems over Europe, interacting with the North African anticyclone and producing a strong western flow. It has to be added also that local uplifting might be reinforced by local topography (over northwestern Africa or the Greek peninsula, for example).

The manuscript text will be modified accordingly.

Specific Comments 1) Page 2462, line 22: "10-15% lower" should be "10-15% higher". (The correction will be inserted in the manuscript)

2) Page 2462 line 24-25: It is stated that the "the relative humidity levels inside the boundary layer are higher than the corresponding lower troposphere level." However, the plot for Antalya in Fig. 1b shows a maximum in relative humidity at 3km.

The mentioned phrase is a general statement, which is clearly applicable for Cairo and Tel-Aviv and to a certain extent for Heraklion and Rhodes, when examining the average vertical summer profiles of relative humidity (Fig. 1). For Antalya, the relative humidity maximum observed around 3 km is probably due to the local topography characteristics (high mountains in the vicinity) in combination with the prevailing atmospheric circulation, as discussed in previous comments.

3) Page 2468, lines 25-29: I do not understand the explanation given here. Why would the high boundary layer mixing heights lead to higher ozone in the boundary layer?

Generally the tropospheric ozone concentrations increase with increasing altitude (Fig. 1). The argument is that the boundary layer ozone concentrations might increase from the influence of the higher ozone values existing at the upper layers. The influence of the boundary layer from the upper reservoir layers is expected to be maximized during the mid-day and the afternoon hours, when the daily maximum of the boundary layer occurs, caused by the thermal instability, transporting down to the surface ozone-rich air masses located above.

4) Page 2469, line 1: Change "tapping" to "trapping". (The correction will be inserted in the manuscript)

5) Page 2470, lines 4-5: "From this analysis it turns out a key factor leading to high upper tropospheric ozone values in the Eastern Mediterranean is the anticylonic influence."

This was not demonstrated in the manuscript. Other studies have shown this, but it is not clear to me how this analysis linked high upper tropospheric ozone to anticyclonic conditions.

In fact, instead of the "lower tropospheric ozone", on which the whole argumentation of the paper was based, it was written by mistake "upper tropospheric ozone", which will be corrected in the manuscript.

6) Page 2471, lines 1-7: Please explain how the CAPE and CIN values are calculated.

As also reported above, responding to a comment of the previous Reviewer, based on our analysis the tropospheric stability or instability atmospheric conditions associated with highest or lowest tropospheric ozone concern in principle extended air masses covering large geographical areas. So, the most important information on CAPE and CINS parameters of the selected days could come from the daily data vertical sounding stations of the Eastern Mediterranean Basin. The CAPE and CINS calculations based on these data are provided by the University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html). As mentioned in the paper, CAPE and CINS values indicating instability of air masses over the area during low ozone days and stability during high ozone days have been observed. In addition, we calculated CAPE and CINS values based on the MOZAIC data using the general literature formulas, which in fact verified the same pattern.

7) Page 2471, line 28: The statement "ozone values in Rhodes, Heraklion and Antalya are inferior to the corresponding in Cairo: : :" is unclear.

As noted in the beginning of the paragraph, this phrase refers to the average summer ozone profiles in all examined airports, presented in Figure 1.

8) Figures 1, 2, 4, 6, 8, and 10 are too small. Perhaps a four-panel plot for each consisting of two rows and 2 columns might provide more space for the individual panels, so that it will be easier to read the plots.

We could try the proposed figure format in the revised manuscript. We think though that the current format has the advantage of an easier visual comparison of the changes of the chemical and meteorological parameters at the different altitudes.