

1 **Reviewer 1 Comment**

2

3 From my initial read of the text, this manuscript has the potential to be an interesting
4 paper in ACP, but unfortunately the presentation of the figures is so poor that I am unable
5 to evaluate the analysis. The authors will have to completely revise and condense
6 their figures before I can make a recommendation to the editor.

7 This paper has 21 figures with a total of 133 panels, 90% of which are too small to
8 be read. This is a result of the authors' poor formatting and visual display, coupled with
9 ACPD's inadequate page layout which routinely makes figures far too small (I complain
10 about this often). Any ACPD paper should be legible when printed onto paper, but for
11 this paper, I can only make out the text in the figures by literally blowing up each panel
12 on my laptop to 300% and then peering very hard to make sure I understand the highly
13 pixelated text. I am not prepared to review a paper such as this when there are over
14 100 tiny panels.

15

16 I also find the figure production to be below the standards of the journal. The authors
17 did not produce the figures of the meteorological fields. All they did was go to the
18 website of the NOAA/ESRL Physical Sciences Division and use the web-based plotting
19 tool to make figures using coarse resolution NCEP reanalyses. While the figures from
20 this website are fine for discussion of a paper when it is in draft form, they are not
21 adequate for ACPD. The text is far too small and the color schemes are terrible and
22 very difficult to interpret. This website also produces the data for each plot in a simple
23 text file that can be used by the authors to plot the figures in a more visually appealing
24 and understandable format, but they chose not to do so. Why did the authors choose
25 to use the NCEP 2.5 degree data to investigate regional scale transport patterns over
26 the Mediterranean? As EU scientists these authors have free access to the superior
27 ECMWF data, which are available at much higher resolution, far more appropriate to
28 this regional scale study. The authors need to switch to ECMWF data and plot it with
29 a resolution of at least 1 degree, preferably with a resolution of 0.5 degrees, if they are
30 to properly understand the regional scale transport patterns. This is especially true for
31 vertical velocity which is essentially useless at 2.5 degree resolution.

32 I have the same criticisms of the HYSPLIT trajectories. The figures were not produced
33 by the authors but simply taken from the HYSPLIT interactive webpage. When they
34 were pasted into the manuscript they were shrunk vertically so that the aspect ratio is

1 all wrong. The authors need to get the trajectory data from the website and plot the
2 trajectories in a legible format.
3 I have similar complaints about the rest of the figures, except for figures 1, 20 and 21.
4 When the authors revise their figures they also need to greatly reduce the number of
5 panels. No one wants to sit down with a paper and read through over 130 panels.
6
7

8 **Authors Response (in Italics)**

9
10 *We would like to thank the reviewer for his comments, which help improving the paper.*
11

12 *At first, we would like to express our surprise for the strong criticism of the reviewer towards*
13 *NOAA/ESRL charts as we had already used them in the same form previously, without any*
14 *objection, in at least 4 papers, which are included in the list of references: 2 in Atmospheric*
15 *Chemistry and Physics (Kalabokas et al., 2007; 2013), 1 in Atmospheric Environment*
16 *(Kalabokas et al., 2008) and 1 in Tellus B (Kalabokas et al., 2015).*
17

18 *Following the reviewer's suggestion, we tried to replace the NOAA/ESRL maps to the*
19 *corresponding ECMWF maps. It appears though that the ECMWF web-page does not offer*
20 *corresponding possibilities for producing composite charts. So, we relied to software*
21 *developed at the LISA-Paris laboratory for the production of Figures based on ECMWF data.*
22

23 *It turned out also that we could not produce temperature anomalies and specific humidity*
24 *anomalies based on ECMWF data. It was possible to produce composite charts of*
25 *geopotential height, wind speed and omega at the 900 and 800 hPa pressure levels, shown in*
26 *new Figs 3 and 8 for the April and May episodes respectively. In the new Figs 2 and 7 the*
27 *composite charts of all examined meteorological parameters are presented at the 850hPa*
28 *pressure level, based on NOAA/ESRL data, but they were plotted by using the same plotting*
29 *tool as for the ECMWF maps, which was the initial suggestion of the reviewer. As observed,*
30 *both chart types (ECMWF and NOAA/ESRL) show more or less the same patterns, which*
31 *suggests that for the examination of high ozone episodes the choice of meteorological map*
32 *type does not influence the used argumentation.*

33 *In any case, the original NOAA/ESRL plotting format does not appear in the main Figure list,*
34 *according to the reviewer's suggestion..*
35

36 *We also improved the layout of the HYSPLIT back-trajectories by using the maximum*
37 *available plot resolution from the web-page, since we do not dispose another alternative.*

38 *We also corrected the aspect ratio of these charts following the reviewer's suggestion.*
39

40 *We also proceeded to a substantial reduction of Figs, as it was suggested by the reviewer.*
41 *With these arrangements the number of Figs in the Figure list is reduced to 11 and the*

1 *number of panels is reduced to 52 (more than 50 % reduction). We also decreased the*
2 *number of panels per page for easier reading. On the other hand, we just created an Annex*
3 *(as it was also suggested by reviewer 2) for shifting there some of the Figs, which might be*
4 *interesting for some readers.*

5

6 *Following the above arrangements the paper has been restructured, as seen in the attached*
7 *revised version, and we believe that the reviewer's requirements are now fully fulfilled.*

8 *The corresponding changes in the revised manuscript are highlighted in yellow color.*

9

10

1 **Reviewer 2 Comment**

2

3 The paper highlights one of the important factors, i.e. synoptic meteorological system,
4 controlling high ozone concentration episodes over Western Mediterranean (W-MED)
5 and Central Europe (C-EU), during spring. The text is generally well written and clear.

6 The time series plots of the observations clearly confirm there were two episodes in
7 which ozone builds up during late April and early May in 2008 over most of the
8 Mediterranean countries. However, when it comes to the proof of the argument, there are a
9 number of statements and images repeating the same messages. The analysis of various

10 parameters that are generated from different types of observations and models

11 is a great idea, but unfortunately they are not always univocal. Therefore I think this

12 paper needs a major revision. Particularly, I would encourage the authors to avoid

13 misinterpreting the results of multiple sources.

14

15 The major concern I have with this paper, particularly in the result and discussion
16 section, is that there is no clear focus on the two regions which are mentioned in the
17 title (i.e. W-MED and C-EU). The NCEP/NCAR reanalysis maps (geopotential height,
18 etc.) include enough evidences to confirm the existence of subsidence over those (two)
19 regions during late April and early May, respectively. Also, there is a positive signal in
20 all selected meteorological parameters on episode days; however the signal is not as
21 strong as expected for some of the parameters. I cannot understand why the authors
22 avoid focusing on them. Furthermore, the mechanisms leading to ozone enhancement
23 as a consequence of high pressure systems should be explained in more detail. One
24 mechanism could be the accumulation of surface ozone which is produced through
25 chemical reactions due to the stagnant air flow. Another one could be linked to ozone
26 flux from the upper troposphere to the surface, which the authors have already tried to
27 prove but without sufficient arguments.

28

29

30 **Authors Response (in Italics)**

31

32 *We would like to thank the reviewer for his comments, which help improving the paper. We*
33 *think that it would be more practical to structure our response, presented below, according to*
34 *the very detailed comments of the reviewer.*

35 *The corresponding changes in the revised manuscript are highlighted in green color.*

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38

Detailed comments:

1) Abstract: “ozone measurement from countries surrounding Western Mediterranean. . .”_The promising title indicates that the study is focused on two different regions, W-MED and C-EU. Then in the abstract the focus changes to only one of them i.e. W-MED. Even the time series are plotted just for W-MED, why? According to the EEA-AirBase maps, Fig.18, the second episode is located over C-EU, isn’t it? Wouldn’t it be better to select a few stations from C-EU and plot their time series for them (as it has been done for W-MED)?

In fact the region of study is the W-MED and the corresponding EMEP stations in that region have been selected for both episodes with highest ozone values over the region. At a later stage of the analysis it came out that especially for the May episode high ozone levels have been also recorded at Central Europe at the same time and for that reason this region was added also to the title. We could add another Figure presenting measurements from some selected Central European stations but this would be in conflict with the remarks of Reviewer 1 who suggests to reduce substantially the number of Fig and which will be reduced to the half of their original number. In addition, the presented EMEP stations in Switzerland and France cover also some parts of the central European domain. For this purpose, also, we will add in the text the stations names as well as geographical coordinates. As a final precaution and for avoiding confusion, we will remove form the title “Central Europe” as the paper is essentially concentrating on the W-MED.

2) Abstract: “the results show that high ozone . . .”_I think here you mean the results of the observation, don’t you?

Yes, indeed. The phrase will be modified accordingly (“results” will be replaced by “observations”).

3) Abstract: “over these areas, strong...”_I think it is too much detail for the abstract section.

This part of the abstract will be reduced according to the reviewer’s suggestion by removing 3 lines from the abstract text (“Over these areas.....500hPa pressure levels”).

4) Introduction: in general the strongest focus of this part is over E-MED region during

1 summer.

2

3 *Yes, indeed. As mentioned, our initial focus was the Mediterranean basin where the highest*
4 *ozone observations are observed in its Eastern part during summer and, in fact, a major*
5 *finding of this paper is that comparable synoptic meteorological conditions exist between*
6 *EMED in summer and WMED in spring during ozone episodes. According to the reviewer's*
7 *suggestion, some more elements on previous studies on the WMED will be added.*

8

9 5) Introduction: "transport times are typically shorter. . ."_wouldn't it be better to
10 rephrase this sentences to something like "they can be transported over longer distances
11 than that in the boundary layer"?

12

13 *This phrase will be modified according to the reviewer's suggestion.*

14

15 6) Introduction: are there any references regarding the frequent existence of anticyclone
16 condition over MED during spring?

17

18 *When examining the average seasonal climatological charts of geopotential heights, it is*
19 *clear that the N. African anticyclone is progressively extending and moving towards the*
20 *Central Mediterranean when passing from winter to spring and summer months*
21 *(<http://www.esrl.noaa.gov/psd/cgi-bin/data/composites/printpage.pl>).*

22

23 7) Data and methodology: I think it would be nice to add the path (precise address) of
24 the data center or website which the data are taken from.

25

26 *The precise address of the data center or website from which the data are taken will be added*
27 *in the text, according to the reviewer's suggestion*
28 *(<http://www.nilu.no/projects/ccc/onlinedata/ozone/>).*

29

30 8) Data and methodology: what is the horizontal and vertical resolution of the
31 CHIMERE model?

32

33 *The current configuration of the CHIMERE model uses a horizontal resolution of 0.25° x*
34 *0.25° and 30 hybrid (σ, p) vertical levels are used to describe the whole troposphere (i.e from*
35 *the ground to 200hPa).*

36

37 9) Data and methodology: the description of methodology is insufficient.

38

1 *An effort will be made to improve the methodology section and for that purpose a sentence*
2 *will be added at the end of this section (“The satellite.....boundary layer”).*

3

4 10) Results and discussion, section 3.1: as it has been mentioned in this part, Fig. 1
5 (lower panel) shows a very good agreement between the results of different stations.
6 So I cannot see the reason of having two different time series plots in Fig. 1?

7

8 *Just to show that the regional ozone episodes affect more WMED countries (Spain +Malta),*
9 *covering most of the basin. It would be difficult to show that without Fig.1a. Especially for*
10 *the April episode, Malta shows the highest ozone concentrations over several days, attributed*
11 *mainly to tropospheric transport.*

12

13 11) Results and discussion, section 3.1: the title of this section is “. . . over Western
14 Mediterranean”, but the description mixes both episodes together. I would strongly
15 recommend the authors to create a separate time series for each episode in each
16 region as it has been done by airbase maps (lowest panel of Fig. 9 and Fig. 18).
17 These maps and time series are enough proof for the confirmation of the existence of
18 two episodes over these regions.

19

20 *At first, as mentioned also above, “Central Europe” will be removed from the title as the*
21 *paper is mainly focused on the Western Mediterranean ozone episodes although Central*
22 *Europe is in fact also influenced at the same time, especially during the May episode.*

23 *The selected April and May episodes seem to be the most characteristic of the season but we*
24 *think that it is worth it to show also the other high (or low) ozone episodes occurring during*
25 *spring 2008 and showing that mid-day ozone concentrations might have the same variation*
26 *pattern even at large distances between rural ozone stations, which indicates regional*
27 *episodes. In addition, due to strong objections from reviewer 1 we need to reduce*
28 *substantially the number of Figs, which have been reduced by about the half of their initial*
29 *number.*

30

31 12) Results and discussion, section 3.2: since there are too many plots, I would suggest
32 the authors to keep the plots related to the episode days in the main paper and
33 move the others (i.e. the plots which they are related to the a few days before episodes)
34 to the supplementary.

35

36 *It will be done so, following also corresponding remarks of reviewer 1, as about half of the*
37 *plots will be moved to the Annex.*

38

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37

13) Results and discussion, section 3.2, a): is there any necessity to explain the low pressure system over another region i.e. E. Europe? How this system leads to high ozone concentration over W-MED?

In fact one of the main points of the paper is that the interaction of high and low pressure systems create conditions of subsidence, especially at the interface of both meteorological systems. So, the extent and the intensity of high and low pressure systems are very important for a better understanding of this phenomenon. A key point is that the WMED area is influenced by this process as the air masses arriving there originate from the subsidence area located in E. Europe.

14) Results and discussion, section 3.2, b): why do the authors describe negative specific humidity over Atlantic or etc.? The main focus of this part must be over WMED and there is strong signal of negative specific humidity over this region (in the lowest panel of Fig. 2), why don't the authors concentrate on that?

Essentially for the same reasons as in the previous remark. The extended subsidence over the Atlantic affects WMED through transport as back-trajectories and the meteorological charts show.

15) Results and discussion, section 3.2, c): do we really need different maps of omega and its anomaly? In fact, both of them have the same messages.

We wanted to put more emphasis that during the ozone episodes, unusual vertical exchange conditions occur (positive omega, indicating subsidence). The omega anomalies could be removed from the main Figure list, also in the spirit of the remarks of reviewer 1 for reducing the total number of Figs.

16) Results and discussion, section 3.2, d): Yes, indeed there is a strong westerly wind toward W-MED a few days before the episode. {It may transfer ozone and its precursors from other places such as eastern US, etc. towards this region (via long-range transport), but there is not enough evidence for that through this maps.} However, on the 26th and 27th of April (in the lowest panel of the left column in Fig. 4), there is a weak wind flow over W-MED due to the existence of a high pressure system.

1 *The back-trajectories in combination with the IASI satellite measurements show that the flow*
2 *over W-MED originates from the high tropospheric ozone area over N. Atlantic. Of course,*
3 *photochemical ozone production inside the anticyclone might also occur, which will be more*
4 *emphasized in the paper.*

5

6 17) Results and discussion, section 3.2, e): in those episode days, there is a positive
7 temperature anomaly over W-MED due to high pressure system. It leads to even more
8 ozone production through photochemical reactions, doesn't it?

9

10 *Yes, of course. But the simultaneous appearance of positive and negative temperature*
11 *anomalies is also a sign of tropospheric processes leading to subsidence occurring at the*
12 *interface of both areas but shifted somewhat towards the negative temperature anomalies*
13 *area, which is associated with colder and richer in ozone tropospheric air masses.*

14

15 18) Results and discussion, section 3.2: “overall, over the same area of subsidence
16 . . .”_how is it possible to have strong subsidence and strong wind together at the same
17 time? Strong wind may even lead to a reduction of ozone by transferring them to the
18 other regions.

19

20 *This is actually a very good point, which might help clarifying the discussion. As mentioned*
21 *also above, during high ozone episodes, we might have strong downward transport (or*
22 *subsidence) and strong winds at the same time, which usually originate from high*
23 *tropospheric ozone reservoirs (a frequently occurring situation observed at the eastern*
24 *Mediterranean during summertime). This is, in fact one of the main points of this paper:*
25 *during high springtime ozone episodes in the WMED comparable synoptic conditions and*
26 *atmospheric processes occur as during high ozone in the EMED in summertime. On the*
27 *contrary, lower ozone levels occur in the troposphere during autumn and winter seasons*
28 *under similar conditions, as the tropospheric ozone levels are significantly lower. In addition,*
29 *in EMED during summer strong westerly winds, transporting boundary layer air from the*
30 *Atlantic, are usually associated with low ozone but the corresponding synoptic conditions are*
31 *quite different if compared with the high ozone episodes associated with strong northerly*
32 *descending winds over the Aegean Sea and the EMED (Kalabokas et al., 2013; Kalabokas et*
33 *al., 2015).*

34

35 19) Results and discussion, section 3.2: “In figure 5, the composite ozone IASI . . .”_ as
36 the authors have already mentioned in this section, there is a signal of high ozone at
37 free troposphere over C-MED and Atlantic. There is no explanation of how these are
38 connected to high surface ozone over W-MED.

39

40 *As also mentioned previously and according to observations, the connection might occur*
41 *through advection, which follows the subsidence observed over the Atlantic. In fact, the*

1 *CMED high ozone maximum observed by IASI is associated with processes occurring within*
2 *the low-pressure system, leading to the enhancement of ozone levels. We agree that according*
3 *to the meteorological analysis no influence is observed from the CMED tropospheric ozone*
4 *maximum to the surface ozone observations in WMED.*

5
6 20) Results and discussion, section 3.2, page 9, first paragraph: I think, adding Fig.
7 7 is just overemphasizing the same messages which have been already explained in
8 Fig. 1, 2, and 3.

9
10 *The figure will be removed following the reviewer's suggestion and will be put in the Annex.*
11 *The idea is just to show that similar synoptic patterns and associated processes occur also at*
12 *700hPa and 500 hPa levels, which indicate deep subsidence throughout the troposphere.*

13
14 21) Results and discussion, section 3.2, page 9, second paragraph: the CHIMERE
15 simulation shows more or less the same results as IASI satellite data. There is high
16 ozone in the free troposphere over C-MED. I cannot understand how this information
17 is connected to high surface ozone over W-MED? I do not recommend the authors to
18 apply a model simulation in this study without any evaluation of that.

19
20 *As mentioned previously, the analysis shows that surface ozone in W-MED is influenced from*
21 *the high ozone reservoir over the Atlantic region (in addition to the photochemical production*
22 *during the last days of the episode). On the contrary, we agree that the high tropospheric*
23 *ozone over C-MED (related to the low pressure system to the east) does not influence the*
24 *surface ozone concentrations in W-MED, as also mentioned above.*

25
26 22) More or less the same recommendations as above are valid for section 3.3.

27
28 *The May episode described in section 3.3 is quite different than the April episode regarding*
29 *synoptic meteorological conditions, as the main anticyclone associated with the ozone*
30 *episode is located in central and northern Europe and the corresponding discussion*
31 *concerning the synoptic influence on ozone levels has been adopted accordingly.*

32
33 23) Conclusions: “in this paper, the investigation of the regional . . .”_what does ‘surrounding
34 countries’ mean? Does it mean C-EU?

35
36 *The phrase means ‘surrounding countries’ of the northern and eastern part of the western*
37 *Mediterranean basin, from which the results of the EMEP stations are presented in Fig. 1*
38 *(Spain, France, Switzerland, Italy, Malta).*

1 24) Conclusions: paragraph 4: how do negative temperature anomaly and strong wind
2 contribute to the high ozone level?

3

4 *As mentioned also previously, strong winds might be associated with high ozone if they*
5 *originate from a high ozone reservoir located in the upper tropospheric layers (as IASI*
6 *measurements indicate). This rapid downward transport, transporting colder and richer in*
7 *ozone air from upper layers located to the north, is associated in fact with a negative*
8 *temperature anomaly, which usually appears next to a positive temperature anomaly area,*
9 *located inside the anticyclone during subsidence conditions. These observations are in*
10 *agreement and support the corresponding observations from the analysis of other*
11 *meteorological parameters, indicating strong vertical tropospheric transport originating from*
12 *northern directions.*

13

14 25) I would strongly recommend the authors to use a larger size for labels, title, etc. for
15 all figures to make them readable.

16

17 *The new ECMWF Figs will take into account the above remarks.*

18

19 Figure 1: The lower time series plot clearly shows the episodes. There is no need to
20 keep both plots. Furthermore, the unit of ozone in the legend should be “ppb” instead
21 of “ug/m3”, shouldn’t it?

22

23 *The figure just shows that the high ozone concentrations occur also at Spain and Malta,*
24 *especially for the April episode, which is important to show its extend over the western*
25 *Mediterranean basin.*

26 *Yes, indeed, “ppb” should be used instead of “ug/m3”, we apologize for this mistake.*

27

28 Figure 2: The right column is specific humidity anomaly, but in the legend it is written
29 specific humidity. It would be recommendable to add the unit of this parameter.

30

31 *In fact, “anomaly” should be added to “specific humidity”. Also, the unit of this parameter*
32 *will be added in the legend.*

33

34 Figure 3: Both omega and omega anomaly have the same messages; I would recommend
35 the authors to keep only one of them. Units are missing.

36

37 *As mentioned also above, only omega will be retained in the main Figure list.*

38

1 Figure 4: Again, adding units would be recommendable.

2

3 *Units will be added in the legend, according to reviewer's suggestion.*

4

5 Figure 9: It is hard to see the white contour over CHIMERE maps. In the legend the
6 color of contours is labeled black instead of white. I do not know what the aim is of
7 putting surface ozone (from EEA-AirBase) maps separately below CHIMERE simulation
8 maps.

9

10 *The white tracer contours of the CHIMERE maps will be replaced by black ones. Also the*
11 *EEA-Airbase observations will be put together with the IASI measurements in the main*
12 *Figure list (new Figs 3, 7).*

13

14

1 **Interactive comment 3.**

2

3 The paper by Kalabokas et al. strongly relies on ozone observations from various
4 measurement networks and platforms but none of the data providers is properly accredited in
5 the acknowledgements. I am part of the operators team of the Swiss National Air Pollution
6 Monitoring Network – which is one of the providers of the EMEP data used here, namely
7 Payerne (CH02) and Chaumont (CH04), even though I am not directly involved in any of the
8 used observations. In other words, I am not writing to claim any personal credits. However,
9 our network’s funding partly relies on a sound annual reporting proofing the interest in our
10 observations by the (scientific) community. Therefore, a proper acknowledgement in
11 publications using our data is vital for the long-term operation of the measurements. This is
12 also true for many other monitoring networks. As host of the WMO/GAW Quality Assurance
13 / Science Activity Centre Switzerland (QA/SACCH), we also continuously promote the
14 timely submission of data to the international data repositories. Missing credit for data made
15 publicly available is one of the main arguments against a (fast) provision of the time series.
16 Thus, the missing declaration of the data source will also jeopardize similar analysis in the
17 long run. Moreover, the EMEP/EBAS data policy accessibly on the EBAS webpage clearly
18 reads: “For scientific purposes, access to these data is unlimited and provided without charge.
19 By their use you accept that an offer of co-authorship will be made through personal contact
20 with the data providers or owners whenever substantial use is made of their data. In all cases,
21 an acknowledgement must be made to the data providers or owners and to
22 the project name when these data are used within a publication.”

23

24

25 **Authors Response (in Italics)**

26

27

28 *We really apologize for the lack of particular acknowledgement to the operators of the air*
29 *quality stations, thinking that mentioning the EMEP programme and its data platform would*
30 *be sufficient. We agree totally with the spirit of this comment. So, for that purpose, we will*
31 *add at first a reference for the EMEP program (EMEP, Chemical Coordinated Center,*
32 *www.emep.int) as well as the station names and their geographical coordinates in the*
33 *manuscript text (Data and Methodology section).*

34 *Finally, we will add the following sentence at the end of acknowledgements. “Our particular*
35 *thanks go to the operators of the following air quality stations: Cabo de Creus in Spain,*
36 *Morvan and Montandon in France, Payerne and Chaumont in Switzerland, Montelibretti and*
37 *Ispira in Italy and Giordan Lighthouse in Malta”.*

38

39 *The corresponding changes in the revised manuscript are highlighted in blue color.*

40

41

42

43

An investigation on the origin of regional spring time ozone episodes in the Western Mediterranean

Pavlos Kalabokas^{1,2}, Jens Hjorth², Gilles Foret³, Gaëlle Dufour³, Maxim Eremenko³, Guillaume Siour³, Juan Cuesta³, Matthias Beekmann³

¹Academy of Athens, Research Center for Atmospheric Physics and Climatology, Athens, Greece

²European Commission, Joint Research Centre (JRC), Institute for Environment and Sustainability (IES), Air and Climate Unit, I - 21027 Ispra (VA), Italy

³Laboratoire Inter-universitaire des Systèmes Atmosphériques (LISA), Universités Paris-Est Créteil et Paris Diderot, CNRS, Créteil, France

Correspondence to: Pavlos Kalabokas (pkalabokas@academyofathens.gr)

Abstract. For the identification of regional spring time ozone episodes, rural EMEP ozone measurements from countries surrounding the Western Mediterranean (Spain, France, Switzerland, Italy, Malta) have been examined with emphasis on periods of high ozone concentrations, according to the variation of the daily afternoon (12:00 – 18:00) ozone values. For two selected high ozone episodes in April and May 2008, composite NCEP/NCAR reanalysis maps of various meteorological parameters and/or their anomalies (geopotential height, specific humidity, vertical wind velocity omega, vector wind speed and temperature) at various tropospheric pressure levels have been examined together with the corresponding satellite IASI ozone measurements (at 3 and 10 km), CHIMERE simulations, vertical ozone soundings and HYSPLIT back trajectories. The observations show that high ozone values are detected in several countries simultaneously over several days. Also, the examined spring ozone episodes over the Western Mediterranean and in Central Europe are linked to synoptic meteorological conditions very similar to those recently observed in summertime ozone episodes over the Eastern Mediterranean (Kalabokas et al., ACP, 2013; Doche et al., ACP, 2014; Kalabokas et al., TellusB, 2015), where the transport of tropospheric ozone-rich air masses through atmospheric subsidence influences significantly the boundary layer and surface ozone concentrations. In particular, the geographic areas with observed tropospheric subsidence seem to be the transition regions between high pressure and low pressure systems. During the surface ozone episodes IASI satellite measurements show extended areas of high ozone in the lower and upper troposphere over the low pressure system areas, adjacent to the anticyclones, which influence significantly the boundary layer and surface ozone concentrations within the anticyclones by subsidence and advection in addition to the photochemically produced ozone there, resulting in exceedances of the 60 ppb standard.

1 Introduction

Surface ozone is a pollutant harmful to both human health and vegetation (Levy et al., 2001; Fuhrer, 2009). Further, in the upper troposphere ozone acts as a powerful greenhouse gas (IPCC, 2007). The concentrations of ozone throughout the troposphere depend on the meteorological conditions driving vertical and horizontal transport and on photochemical ozone production from its precursors, nitrogen oxides (NO_x) volatile organic

1 compounds (VOCs) and carbon monoxide (Delmas et al., 2005; Seinfeld and Pandis, 2006; Monks et al., 2015).
2 **Recent model studies and studies based on observational constraints indicate that a little less than 90% of the**
3 **ozone found in the troposphere is formed photochemically within the troposphere, the remaining part is brought**
4 **down to the troposphere by stratosphere-troposphere exchange (Monks et al., 2015).**

5 The European network of surface ozone monitoring stations shows persistently exceedances of the European
6 long-term target value for protection of human health, under anticyclonic synoptic meteorological conditions
7 during the warm season in southern and central Europe (EEA, 2015). The Mediterranean area is particularly
8 exposed to ozone pollution because of the combination of the specific meteorological conditions prevailing
9 during spring and summer and the regional air pollutant emissions. Data collected from air pollution monitoring
10 stations in combination with results of measurement campaigns show that ozone concentrations in the
11 Mediterranean Basin are relatively high: Lelieveld et al. (2002) found that summer ozone concentrations over the
12 Mediterranean are a factor of 2.5–3 higher than in the hemispheric background troposphere, in the boundary
13 layer and up to 4 km altitude. Rural stations in continental Greece, Italy, Malta and eastern Spain report summer
14 average ozone values of about 60–70 ppbv, significantly higher than values in Northern and Western Europe
15 (Bonasoni et al., 2000; Kalabokas et al., 2000; Millan et al., 2000; Kourtidis et al., 2002; Kouvarakis et al., 2002;
16 Nolle et al., 2002; Kalabokas and Repapis, 2004; Paoletti, 2006; Sánchez et al., 2008; Schürmann et al., 2009;
17 Velchev et al., 2011, Kalabokas et al., 2008; Kleanthous et al., 2014; Cristofanelli et al., 2015). Results from 3-
18 D chemistry transport models also suggest that ozone concentrations are higher than for the rest of Europe (e.g.
19 Johnson et al., 2001). High ozone values in the Mediterranean are typical not only for ground level
20 measurements, but for the entire boundary layer as well as the entire lower troposphere (Millan et al., 1997,
21 2000; Kalabokas et al., 2007).

22 The Mediterranean climate with frequent anticyclonic, clear sky conditions in spring and summer favours
23 photochemical ozone formation in the troposphere. Furthermore, Mediterranean tropospheric ozone levels are
24 influenced by long-range transport of ozone and its precursors from Europe, Asia and even North America as
25 well as emissions of precursors from sources around the Basin, particularly the large cities (Lelieveld et al.,
26 2002, Gerasopoulos, 2005, Safieddine et al., 2014). Also natural VOC emissions in the area have been found to
27 be important ozone precursors (Richards et al., 2013). **Several studies in the Western part of the Mediterranean**
28 **involving measurements as well as model simulations (e.g. Millan et al., 1997, 2000, Querol et al. 2016) have**
29 **addressed the causes of episodes with high ozone concentrations. It was found that the typical synoptic**
30 **meteorological conditions found during the summer in this part of the Mediterranean, with a lack of strong**
31 **synoptic advection combined with the orographic characteristics and the sea and land breezes favour episodes**
32 **where high levels of ozone are accumulated by recirculation of air masses loaded with ozone precursors.**

33 Anticyclones are generally linked to atmospheric subsidence, which seems to be particularly important over the
34 Eastern Mediterranean as a cause of elevated ozone concentrations. During the summer period, the
35 Mediterranean area is directly under the descending branch of the Hadley circulation, caused by deep convection
36 in the tropics (Lelieveld, 2009). However a main reason for the strong subsidence observed in the Mediterranean
37 Basin appears to be an impact of the Indian monsoon, inducing a Rossby wave that by the interaction with the
38 midlatitude westerlies produces adiabatic descent in the area (Rodwell and Hoskins, 1996, 2001; Tyrllis et al.,
39 2013). In general, in the Eastern Mediterranean strong deep subsidence in the lower troposphere influencing the

1 boundary layer has been documented, based essentially on the analysis of MOZAIC vertical ozone profiles as
2 well as surface ozone and satellite measurements (Kalabokas et al., 2007; Kalabokas et al., 2008; Eremenko et
3 al, 2008; Foret et al., 2009; Liu et al., 2009; Kalabokas et al., 2013; Doche et al., 2014; Kalabokas et al., 2015).
4 Also data analysis based on large-scale atmospheric modeling studies (Li et al, 2001; Richards et al., 2013; Zanis
5 et al., 2014; Safieddine et al., 2014; Tyrlis et al., 2014) shows the importance of vertical transport of ozone in the
6 Mediterranean area, particularly in its eastern part.

7 Recent research based on vertical MOZAIC ozone profiles (Kalabokas et al., 2015) suggests that during days
8 with the highest ozone levels for both the free lower troposphere (1.5-5 km) and the boundary layer (0-1.5 km)
9 over the middle-eastern airports of Cairo and Tel-Aviv there are extended regions of strong subsidence in the
10 Eastern Mediterranean but also in Eastern and Northern Europe and over these regions the atmosphere is dryer
11 than average.

12 The influence of tropospheric transport of ozone at hemispheric scale, on surface ozone concentrations, is an
13 issue of potential importance to ozone abatement policies. Vertical transport of ozone is of particular relevance
14 to the ozone transport over long distances in the troposphere because the lifetime of ozone in the free
15 troposphere is longer so it can be transported over longer distances than in the boundary layer (HTAP, 2010).
16 The impact on surface ozone of the long range transport of ozone has been extensively investigated in the US,
17 where a clear difference has been observed between the eastern part, where surface ozone concentrations have
18 decreased very significantly, and the western part, where most sites do not show such a trend, apparently due to
19 the effects of transport (Cooper et al., 2012). Studies based on observations and trajectory modelling indicate
20 that transport above the boundary layer and entrainment of ozone from the free troposphere has an important
21 impact on surface ozone concentrations at several sites in the western US (Cooper et al., 2011; Langford et al.,
22 2015). Also a changing seasonal cycle of ozone, with a tendency towards a maximum earlier in the year, has
23 been tentatively explained by the influence of atmospheric transport patterns combined with a change in the
24 temporal and spatial emissions of ozone precursors (Parrish et al., 2013).

25 The importance of hemispheric transport for Mediterranean air pollution in particular has previously been
26 highlighted in the modelling study by Stohl et al. (2002) of intercontinental transport of air pollutants, who found
27 the highest concentrations of a passive tracer for North American emissions in the Mediterranean Basin. The
28 modelling studies of Richard et al. (2013) and Safieddine et al. (2014) both come to the conclusion that the
29 emission sources within the Mediterranean area have a dominating influence on surface ozone while remote
30 sources, e.g. in Asia and North America, are more important than local sources for ozone concentrations at
31 higher altitudes (above 700 hPa according to Richards et al. 2013). The radiative (climate) impact of ozone
32 depends mainly on the concentrations above the boundary layer. Relatively high ozone concentrations are
33 reported in the Central Mediterranean also during the winter-early spring period (Nolle et al., 2002) while, in an
34 extended observational study in the Western Mediterranean basin a converging trend between the background
35 rural ozone values and the urban ozone values was reported (Sicard et al., 2013).

36 Many studies of Mediterranean ozone have been focused on the Eastern part of the Mediterranean basin where
37 the influence of downwards transport appears to be more important than it is in the Central and Western part,
38 although also at this part of the Mediterranean very frequently anticyclonic conditions dominate in spring and

1 summer months. The mechanisms governing ozone levels in the Western and Central Mediterranean appear to
2 need further clarification particularly regarding the conditions during springtime. For example, in Kalabokas et
3 al. (2008) as well as in the more recent study by Zanis et al. (2014) it was found that rural background ozone
4 monitoring stations in the Western and Central Mediterranean show climatological spring time ozone maximum
5 in April-May while the corresponding rural background ozone Eastern Mediterranean stations, have their
6 climatological maxima in July-August. Also, in Eastern Greece and the Aegean Sea the summer rural
7 background ozone levels are systematically higher than the corresponding ones observed in the Central
8 Mediterranean (Malta) and the Eastern Mediterranean (Cyprus). The above is contrary to the predictions of a
9 model simulation that suggested a rather uniform ozone distribution across the basin (Zanis et al., 2014).
10 Understanding the origin of high ozone levels during spring time is particularly challenging because this is a
11 period where photochemical formation in the troposphere is increasing due to rising sun intensity, but also
12 stratospheric intrusions may be of relevance as these have been found to have a maximum in Southern Europe in
13 spring/early summer (Beekmann et al., 1994; Monks, 2000).

14 The focus of the present study thus is to improve the understanding of ozone behaviour over the Western
15 Mediterranean and the surrounding area to the north part of the basin, towards central Europe in the springtime.
16 In particular, it aims at investigating to which extend surface ozone concentrations during the high ozone
17 episodes are influenced by entrainment of ozone rich air. Further, this study investigates the factors controlling
18 ozone distribution in the free troposphere, particularly transport within the troposphere, stratosphere-troposphere
19 exchange and photochemical formation in the free troposphere. Two episodes in late April and early May have
20 been selected for a detailed analysis. We analyze the two spring time episodes of high surface ozone
21 concentrations over the Western Mediterranean and the surrounding area using a comprehensive combination of
22 surface observations, Infrared Atmospheric Sounding Interferometer (IASI) satellite observations,
23 meteorological maps, back trajectories and regional air quality modelling to understand the principal
24 mechanisms contributing to these events.

25

26 **2 Data and methodology**

27 The following data will be used in the analysis:

28 1) Ozone measurements from the European Monitoring and Evaluation Programme (EMEP) and European
29 Environment Agency (EEA) air pollution network

30 The surface ozone measurements during spring 2008 (March-May) of some EMEP (EMEP, 2015) rural ozone
31 stations (<http://www.mlu.no/projects/ccc/online/ozone/>) surrounding the western Mediterranean basin
32 [Spain, Cabo de Creus (ES10, 42.31 °N, 3.31 °E), France, Morvan (FR10, 47.26 °N, 4.08 °E) and Montandon
33 (FR14, 47.30 °N, 6.83 °E), Switzerland, Payerne (CH02, 46.81 °N, 6.94 °E) and Chaumont (CH04, 47.04 °N,
34 6.97 °E), Italy, Montelibretti (IT01, 42.10 °N, 12.63 °E) and Ispra (IT04, 45.80 °N, 8.63 °E), Malta, Giordan
35 Lighthouse (MT01, 36.07 °N, 14.21 °E)] have been analyzed with emphasis on periods of high ozone, focused
36 mostly on the day-to-day variations of the mean afternoon (12:00 – 18:00) ozone values. In addition, during the
37 selected spring ozone episodes the measurements of the EEA air pollution network are also taken into account
38 for the analysis (EEA, 2015).

1 2) Composite NOAA/ESRL and ECMWF Reanalysis maps

2 Composite or daily NOAA/ESRL and ECMWF reanalysis meteorological maps covering Europe and N. Africa
3 and corresponding to periods of high ozone have been plotted for the following meteorological parameters:
4 Geopotential height, specific humidity anomaly, vertical wind velocity Omega (and anomaly), vector wind speed
5 and temperature anomaly. The examination was focused on the tropospheric pressure levels at 850, 700 and
6 500hPa levels (for space limitations mainly the 850hPa charts are presented). The NOAA/ESRL charts are based
7 on grids of 2.5×2.5 degrees, following the procedure of Kalnay et al. (1996) while the horizontal resolution of
8 ECMWF charts is 0.25×0.25 degrees.

9 3). HYSPLIT back trajectories

10 Six-day back trajectories were calculated with end points at 50, 500 and 1500 meter altitude, using the NOAA
11 HYSPLIT model with GDAS meteorological data (Draxler and Rolph, 2015) for the EMEP stations and the days
12 of the selected ozone episodes. The GDAS data have a horizontal resolution of 1 degree and 23 vertical levels
13 between 1000 and 20 hPa.

14 4). Satellite IASI ozone measurements

15 Satellite observations provide interesting possibilities to support the analysis of ground measurements as well as
16 modeling simulations. Indeed, during the last decade, satellite observations of tropospheric ozone have been
17 developed and have become more and more precise (e.g. Fishman et al., 2003; Liu et al., 2005; Coheur et al.,
18 2005; Worden et al., 2007; Eremenko et al., 2008). These observations are now able to complement in situ
19 observations, offering wide spatial coverage and good horizontal resolution.

20 The IASI instrument (Clerbaux et al., 2009), on board the MetOp-A platform since 19 October 2006, is a nadir-
21 viewing Fourier transform spectrometer operating in the thermal infrared between 645 and 2760 cm^{-1} with an
22 apodized spectral resolution of 0.5 cm^{-1} . The IASI field of view is composed of a 2×2 matrix of pixels with a
23 diameter at nadir of 12 km each. IASI scans the atmosphere with a swath width of 2200 km, allowing the
24 monitoring of atmospheric composition twice a day at any (cloud-free) location. The spectral coverage and the
25 radiometric and spectral performances of IASI allow this instrument to measure the global distribution of several
26 important atmospheric trace gases (e.g. Boynard et al., 2009; George et al., 2009; Clarisse et al., 2011). As in
27 Doche et al. (2014), IASI data at 3 and 10 km height are used here for analysis. These levels are representative
28 for the lower and upper troposphere respectively. However, due to the limited vertical sensitivity and resolution
29 of IASI, ozone concentrations retrieved at 3 km describe the ozone variability from roughly 2 to 8 km, and ozone
30 concentrations retrieved at 10 km describe the ozone variability from 5 to 14 km (Dufour et al., 2010). Despite
31 this overlapping, recent studies show that uncorrelated information from the lower and the upper troposphere can
32 be derived from IASI (Dufour et al., 2010, 2012, 2015).

33 5) Vertical ozone soundings

34 Further information about the vertical ozone distribution is obtained from the ozone soundings made from the
35 site of Payerne, Switzerland, Uccle, Belgium and Hohenpeissenberg, Germany. These are regularly carried out
36 with balloon launches starting at 11 AM UTC using ECC (Electrochemical Concentration Cell) ozonesondes in
37 Payerne and Uccle and Brewer-Mast ozonesondes in Hohenpeissenberg. The data were downloaded from the

1 website of the World Ozone and Ultraviolet Data Centre (WOUDC, 2015), uncertainties on the measured ozone
2 concentrations for ECC ozonesondes are between 5 and 10% (Rene Stübi, personal communication).

3 6). Regional air quality simulations

4 The CHIMERE model (Menuet et al, 2013) is a state-of-the-art model widely used for pollution and air quality
5 studies (Rouil et al., 2009; Beekmann and Vautard, 2010). For the purpose of this study, we have used a version
6 of the model covering a western European domain (35° N – 70° N latitude, 15° W - 35° E longitude). The whole
7 troposphere is described from ground to about 200 hPa using 30 hybrid (sigma, P) levels. The meteorological
8 forcing is given by the IFS forecast of the ECMWF based each day on the 0 and 12 am analysis. The
9 anthropogenic emissions are prescribed by using the TNO inventory (Kuenen et al, 2014) while natural
10 emissions are calculated by the MEGAN module (Guenther et al, 2006). A passive tracer has been used to
11 analyse the dynamics patterns responsible for ozone transport, which is initialized and emitted in the top model
12 layer (11-12 km) i.e within the upper troposphere every hour. A 10- day period (spin-up period) is simulated
13 before each targeted period to establish a kind of equilibrium state of the tracer. Moreover, simulations have also
14 been made by switching off emissions for both periods under study. Differences between simulations with and
15 without emissions is a proxy of the photochemical production of ozone within the boundary layer. **The current
16 configuration of the CHIMERE model uses a horizontal resolution of 0.25° x 0.25° and 30 hybrid (σ,p) vertical
17 levels are used to describe the whole troposphere (i.e from the ground to 200hPa).**

18 The methodology used in this paper is the following:

19 At first, in order to minimize local pollution effects and focus on boundary layer ozone measurements
20 representative of a wider geographical area than the station location, only the afternoon (12:00 – 18:00) ozone
21 concentrations, typically representing a well-mixed boundary layer, have been analyzed.

22 Also, recent observations over the Mediterranean both from MOZAIC tropospheric ozone profiles (Kalabokas et
23 al., 2013) as well as satellite data (Zanis et al., 2014) show a strong anticorrelation between ozone and
24 atmospheric humidity. Therefore, in dry and descending air masses originating from the upper tropospheric
25 layers, higher tropospheric ozone levels would be expected. It is well known that the concentration of water
26 vapour in the troposphere (specific humidity) tends to decrease with increasing altitude because the lower air
27 temperatures at higher altitudes cause elimination of water vapour by condensation followed by precipitation.
28 This makes specific humidity an indicator of subsiding air masses, which will be used in the analysis.

29 In addition, previous research carried out in the Mediterranean (Kalabokas et al., 2007; Kalabokas et al., 2008;
30 Velchev et al., 2011; Kalabokas et al., 2013; Kalabokas et al., 2015) suggested that there is a strong link between
31 synoptic meteorology conditions and ozone concentration variability.

32 Based on the above, a systematic investigation of the composite meteorological maps during two spring high
33 ozone episodes at the 850 hPa pressure level was carried out, until 5 days before the event for the meteorological
34 parameters mentioned previously. The purpose of the analysis is to identify areas of high subsidence in the free
35 troposphere, which could potentially influence the examined surface ozone measurements, considering also that
36 the afternoon (12:00 – 18:00) ozone concentrations are quite representative of the boundary layer values. The
37 high subsidence areas were detected in the first place by the positive vertical velocity omega (and anomalies) as

1 well as the negative specific humidity anomalies. Also, the geographical distributions of geopotential heights,
2 temperature anomalies and vector wind speed were very useful in the examination of the influence of synoptic
3 meteorological conditions on ozone concentrations. In addition, air mass back trajectories (NOAA HYSPLIT),
4 satellite IASI ozone measurements at the 3km and 10km levels, afternoon surface ozone measurements from the
5 EEA European network and vertical ozone profile measurements in Central Europe during a selected ozone
6 episode, were used for the analysis. Finally, CHIMERE tracer simulations and modelling of ozone field for the
7 April and May 2008 ozone episodes have been performed for the validation of the analysis of measurement data.
8 **The satellite tropospheric ozone measurements are used to monitor the movement of the high tropospheric ozone**
9 **reservoirs and their potential influence to the boundary layer.**

11 **3 Results and discussion**

12 **3.1 Surface Ozone measurements in the Western Mediterranean basin in spring 2008**

13 In Fig. 1 (upper panel) the afternoon (12:00 – 18:00) rural ozone concentrations of selected EMEP stations from
14 countries surrounding the Western Mediterranean basin (Spain, France, Switzerland, Italy, Malta) during spring
15 (March – May) 2008 have been plotted. A first investigation of the plots leads to the following remarks:

16 a). Episodic periods of high ozone (and also for low ozone) may last for several days and they can be detected
17 simultaneously in many countries surrounding the western Mediterranean basin.

18 b). The 60 ppb ($120 \mu\text{g}/\text{m}^3$) EU standard for human health protection can be exceeded over many days and in
19 many countries.

20 From the above stations, the ozone concentrations measured at the EMEP stations in France and Switzerland as
21 well as the EMEP station in Northern Italy (IT04, JRC-Ispra), presented in Fig. 1 (lower panel), show generally a
22 very good agreement between each other. This feature is remarkable given their different site characteristics, the
23 large distance between them as well as their location relative to the huge natural barrier of the Alpine
24 mountainous area. So, the high mid-day ozone concentrations observed simultaneously over many countries
25 could be considered as regional ozone episodes. The two regional spring 2008 ozone episodes with the highest
26 ozone concentrations in the examined area were on 26-27 April, 2008 and 7-9 May, 2008; they have been
27 selected for further analysis.

29 **3.2 Geographical distribution of meteorological parameters, IASI tropospheric ozone measurements and** 30 **CHIMERE modelling tracer simulations during the April 26-27, 2008 ozone episode**

31 **The composite NOAA/ESRL Reanalysis maps of geopotential height, vertical velocity omega, vector wind**
32 **speed, temperature anomaly and specific humidity anomaly at 850 hPa as well as IASI satellite ozone**
33 **concentrations for the high ozone episode of April 26-27, 2008 are presented in Fig. 2, while the corresponding**
34 **composite maps for 2, 3 and 5 days before the episode, are shown in Figs I-III of the Annex. In addition, the**
35 **corresponding ECMWF charts at 900hPa and 800hPa for geopotential height, vertical velocity omega and vector**

1 wind speed are shown in Fig. 3. The corresponding composite maps at the 700hPa and 500 hPa pressure levels
2 have also been plotted and analyzed (not shown). The examination of these meteorological charts leads to the
3 following remarks:

4 a). The anticyclone located to the west of the N. African coast is progressively strengthening and expanding after
5 April 23-24, 2008 while moving rapidly towards the European continent. At the same time the low pressure
6 system installed over E. Europe on April 21-24 is progressively reduced to a cut-off low on April 26-27 while
7 moving towards Greece and Turkey.

8 b). Negative specific humidity anomalies are observed all over N. and E. Europe as well as over the Atlantic
9 Ocean while at the peak of the episode (April 26-27) the dry air masses prevail over a vast and extended area
10 from Russia to NW Africa and the Atlantic.

11 c). An extended area of positive vertical velocity omega (and anomalies), which is a direct sign of subsiding air
12 masses, is observed over the W. Mediterranean and the Atlantic and is moving rapidly towards the Central
13 Mediterranean. Five days before the episode (April 21-22) the maximum of subsidence area was observed over
14 the Iberian Peninsula and the adjacent ocean up to the Canary islands. Then it is moving rapidly eastwards,
15 showing a maximum over Italy at the peak of the episode (April 26-27) as well as a secondary maximum over
16 Germany. The positive omega anomalies indicate that the observed subsidence during the episode days is higher
17 than usual for this period of the year and the examined geographical regions.

18 d). A strong westerly flow is observed over the Western Mediterranean region of subsidence with the air masses
19 originating from the region of N. Atlantic where a deep and extended low pressure system is observed. As seen,
20 the extension of the N. African anticyclone towards the Western Mediterranean changes progressively the very
21 strong westerly flow (observed 5 days before the episode, April 21-22) to a strong north-westerly. Also, strong
22 N-NW winds over the Central Mediterranean prevail at the interface area between the cut-off low and the
23 anticyclone.

24 e). Strong and extended negative temperature anomalies are observed 5 days before the episode over the Iberian
25 peninsula and the adjacent N. African coast, moving towards the Central Mediterranean and the Libyan coast.
26 The negative temperature anomaly area becomes more extended during the episode days, indicating that colder
27 atmospheric conditions than the normal prevail during the ozone episode over the corresponding areas, while
28 during photochemical ozone episodes higher than normal air temperatures would be expected. This can be
29 explained as an impact of transport of cold air masses to these areas.

30 Overall, over the same area of subsidence, strong winds together with positive omega vertical velocity (and
31 anomalies) are observed as well as negative humidity anomalies, thus indicating a strong descending air current,
32 which transports rapidly tropospheric air towards the boundary layer.

33 In Figure 1, the composite IASI satellite ozone concentrations at 3km for April 26-27, 2008 are presented as
34 well, while in Fig. IV of the Annex, the composite ozone IASI measurements from April 21, 2008 to April 27,
35 2008 at two altitude levels (3 km and 10 km), which are considered representative for the lower and the upper
36 troposphere (Doche et al., 2014) , even if a partial overlap occurs due to reduced resolution (see section 2), are
37 also shown. An extended area of very high ozone in the upper and lower troposphere is observed over the N.

1 Atlantic over the area covered by the low pressure system. At the same time, in the Central Mediterranean a high
2 tropospheric ozone area is progressively formed at both examined tropospheric layers (3 and 10 km) and having
3 its maximum on April 26-27. This observation could be associated with the information extracted from the
4 analysis of the composite meteorological charts over this Central Mediterranean area where clear indications of
5 very strong subsidence of dry air masses is observed together with very strong north-westerly winds recorded
6 over the Mediterranean at 850hPa (Fig. 1 and Annex Figs I-III), especially for April 26-27. The same features
7 are also observed at the pressure levels of 700hPa and 500 hPa (not shown).

8 For a more detailed analysis during the maximum of the episode (April 26-27), in Fig. 4 the daily IASI satellite
9 ozone measurements at 3 and 10 km altitude are presented in a higher concentration resolution as well as in a
10 more restricted geographical domain (same as the CHIMERE simulation outputs), where the above described
11 characteristics of the geographical distribution of tropospheric ozone appear more clear. Also, for a more
12 detailed investigation of the corresponding meteorological characteristics at higher tropospheric levels, in Annex
13 Fig. V the daily meteorological charts of columnar precipitable water anomaly and geopotential heights at
14 500hPa and 700hPa for April 26 and 27, 2008 are presented. The very extended areas with negative columnar
15 precipitable water anomalies show that the dry conditions, indicating subsidence, prevail over the whole
16 troposphere, mostly over its lower part, over a vast area and especially at the interface areas of the high and low
17 pressure systems at 500 hPa, very similar to what was already observed at 850 hPa (Fig. 2). So, it comes out that
18 during the ozone episode the same synoptic pattern is observed in the troposphere up to 500 hPa (Annex Fig. V)
19 but even at higher levels (not shown). Also in Fig. 3, the graphical representation on the map of the daily
20 evolution of the hourly surface ozone concentrations at 15:00 h as recorded in the air pollution stations of the
21 EEA-AirBase network (EEA, 2015) for 26-27 April shows very high concentrations, exceeding 70 ppb,
22 appearing at many locations in Spain as well as in S. France and NW Italy. As mentioned previously, during the
23 afternoon hours the influence of the free troposphere on the boundary layer is maximized through vertical
24 mixing, and the composite meteorological maps show a large corridor of subsiding dry air masses over the same
25 area (Fig. 1, Annex Figs I-III), which is a very strong indication that the surface ozone concentrations could be
26 influenced by entrainment of subsiding ozone-rich air. Obviously, the distribution of ozone concentrations
27 measured at the ground level stations show important differences from those observed at 3 km height (Fig. 3), as
28 expected, due to the barrier, caused by the temperature inversion, between the boundary layer and the free
29 troposphere and the importance of photochemical formation and transport of ozone within the boundary layer,
30 where the precursor concentrations are relatively high.

31 In order to study in more detail the atmospheric processes prevailing during the examined April ozone episode
32 and complete the analysis of the dynamic atmospheric conditions during the examined events, regarding more
33 specifically the origin of ozone measured in the boundary layer, we have performed simulations with the
34 CHIMERE model using tracers to analyze transport patterns. The results of the CHIMERE simulations for the
35 April episode are shown in Fig. 5, where simulations of the ozone field (together with the iso-contours of the
36 high tropospheric tracer) are presented. Also, in Annex Fig. VI simulations of upper tropospheric tracer
37 concentrations and simulations of photochemical ozone production are shown. As described earlier (cf Chapter
38 2), we use a tracer initialized within the model top layer in the upper troposphere at about 11 km height.
39 Inspecting qualitatively simulated concentrations at 3 km and 1.5 km altitude (Fig. 5), it is shown that the upper

1 tropospheric tracer is present at these levels, indicating significant downward transport of upper troposphere air
2 masses to the boundary layer, which coincides with the maximum concentrations of the CHIMERE simulated
3 ozone field at 3 and 1.5 km covering a large area from Switzerland to Malta (Fig. 5). Combining that with the
4 meteorological charts (Fig. 2), on the outer side of the low-pressure area and at the interface with the N. African
5 anticyclone high values of the upper tropospheric tracer indicating considerable subsidence are observed at 5 km
6 altitude (not shown), which becomes even stronger and more extended at the 3 km and 1.5 km altitudes (Fig.5).
7 So, following the geographical distribution of the upper tropospheric tracer at 3 km and 1.5 km (Fig.5),
8 tropospheric subsidence is observed at the periphery of the anticyclonic area and at the interface with the low-
9 pressure area. Therefore, the downward transport of upper tropospheric ozone is influencing the boundary layer
10 over the examined location. The observed patterns, based on CHIMERE tracer simulations and on ozone
11 concentrations are quite consistent with the observed values of the IASI instrument that show the same structures
12 (Figs 2,4), as well as with the analysis of meteorological parameters based on composite charts (Fig. 2) and
13 described above. Regarding the comparison of the geographical distribution of the CHIMERE ozone field (Fig.
14 5) with the IASI measurements at 3 km or 700hPa (Figs 2, 4), a clear difference over the N. Atlantic and also
15 differences over the Ionian Sea and the Aegean Sea are observed. As discussed above, over these regions the
16 prevailing low pressure systems, inducing lower tropopause heights, are associated with high tropospheric ozone
17 levels. It has to be mentioned that direct comparison of CHIMERE simulations at 3 km with the corresponding
18 IASI measurements presents some weaknesses, especially due to the fact that IASI is sensitive to a height range
19 between 2 and 8 km and the IASI averaging kernels need to be applied to CHIMERE to make it comparable with
20 IASI (Eremenko et al., 2008).

21 The HYSPLIT back-trajectories during the April episode arriving at the EMEP stations in Italy (JRC-Ispra) and
22 on Malta (Fig. 6) show in fact subsiding air masses arriving from northern directions, on April 25 and 26 for
23 Ispra and on April 27 for Malta and give more or less the same picture as the Swiss stations (not shown).

24 The combination of the information from the composite meteorological maps (Fig 2) and the HYSPLIT back-
25 trajectories for the April episode (Fig. 6) shows that the prevailing NW wind during the days preceding the
26 episode passed over the N. Atlantic region, where the IASI satellite detects a high ozone area extended
27 throughout the whole troposphere (Figs 2, 4), which beyond any doubt contributes to the high surface ozone
28 values. So, tropospheric transport of air rich in ozone from northern directions is likely to give an important
29 contribution to the high surface ozone levels through the processes of advection and subsidence.

30 It should be noticed at this point that the interpretation of the back trajectory information is much more efficient
31 if it is done in combination with the examination of the composite meteorological charts (and ideally satellite
32 ozone measurements and modelling simulations). In this way the influence from areas with strong tropospheric
33 subsidence and low atmospheric humidity (indicating high ozone) at the various tropospheric pressure levels
34 could be easier assessed.

35 In summary, the April 26-27, 2008 ozone episode could be considered as a clear case of the tropospheric
36 influence to the boundary layer through ozone transport, which is added to the photochemically produced
37 boundary layer ozone. The analysis of meteorological charts at various tropospheric pressure levels helps in
38 understanding that the strong subsidence occurring over the central Mediterranean plays a key role in explaining

1 this surface ozone episode on April 26-27, 2008. The IASI observations of lower and upper tropospheric ozone,
2 helps in understanding that the subsidence of ozone-rich air masses characterizes this surface ozone episode. The
3 evolution of the above described phenomenon can be effectively monitored by examining the composite
4 meteorological maps at 850hPa during the episode days (as well as at 5-days, 3-days and 2-days before the
5 episode, **Annex Figs I-III**). As mentioned, the examination of the 700hPa and 500hPa pressure levels (not
6 shown) gives comparable results.

7 Based on the above results, the simultaneous strengthening and expansion of the African anticyclone and the
8 formation of a cut-off low over the SE Balkans during the episode seem to give rise to the large scale subsidence,
9 inducing a strong downward transport of cold air masses over the western Mediterranean and the surrounding
10 areas while the maximum intensity of the phenomenon is observed in the geographical area located between the
11 two synoptic atmospheric systems. As mentioned, the signs of stronger tropospheric subsidence during the
12 episode days can be clearly observed from the negative specific humidity anomalies as well as the negative
13 columnar precipitable water anomalies associated with positive omega anomalies (downward motion). The
14 evolution of the temperature anomaly is interesting as the growing region of negative temperature anomaly over
15 more or less the region of subsidence (mostly to its west) as the episode develops, could be explained by the
16 descend of colder air from the upper tropospheric layers, **transported from more northerly latitudes**, caused by
17 the interaction of the low pressure system (located to the east) and the anticyclone (located to the west). This is
18 also consistent with the descent of upper tropospheric tracers simulated with the CHIMERE model. All the
19 above features accompanying a situation with deep subsidence appear in all examined pressure levels at 850 hPa,
20 700hPa and 500 hPa.

22 **3.3 Geographical distribution of meteorological parameters, IASI tropospheric ozone measurements and** 23 **CHIMERE modelling tracer simulations during the May 7-9, 2008 ozone episode**

24 As it will be shown in the next paragraphs, the examination of the May 7-9, 2008 episode leads to comparable
25 remarks regarding synoptic meteorology and geographical distribution of meteorological parameters with the
26 April episode, although in the May episode the meteorological systems and especially the anticyclone are located
27 further to the north. **In Figure 7 the composite NOAA/ESRL Reanalysis maps at 850 hPa and IASI**
28 **measurements at 3 km for the high ozone episode of May 7-9, 2008 are presented. The corresponding composite**
29 **charts at 850 hPa for 2, 3 and 5 days before the episode are presented in Annex Figs VII-X. In addition, the**
30 **corresponding ECMWF charts at 900hPa and 800hPa for geopotential height, wind speed and vertical velocity**
31 **are shown in Fig. 8. Also, the same plots for the 700 hPa, 500 hPa and 300 hPa level have been also created (not**
32 **shown). The examination of all above plots gives the following:**

33 a). A deep and extended low is located over N. Atlantic while a strong anticyclone prevails over N. Europe,
34 centered between Great Britain and Scandinavia. At the same time deep and extended low-pressure systems over
35 the Polar Regions and E. Europe are formed, associated with very dry conditions observed throughout the whole
36 troposphere (**Fig. 7**).

1 b). Over a part of the anticyclonic area and especially at the region of interface of the anticyclone with the low
2 pressure systems to the east, an extended area of positive vertical velocity omega (and anomalies) is observed,
3 associated also with extended areas of low-humidity air masses (Fig. 7), which are clear signs of strong
4 subsidence lasting for many days. The signs of subsidence are particularly strong also at the higher tropospheric
5 pressure levels up to 300hPa (not shown) indicating the occurrence of a large-scale tropospheric phenomenon.

6 c). A strong contrast is observed (Fig. 7) between positive and negative temperature anomalies in W. and E.
7 Europe respectively (and also geopotential height anomalies, not shown), which is similar to what has been
8 observed for the April episode, showing strong influence of transport of air masses.

9 d). Stagnant conditions are observed at the center of the N. European anticyclone located over the North Sea
10 while at the southern part of the anticyclone a strong positive vertical omega velocity is observed (indicating
11 subsidence), which is associated with a stronger vector wind velocity over C. Europe. In fact, a strong air current
12 starting from the N. Atlantic makes a circular clockwise motion at the periphery of the anticyclone towards the
13 East while the flow at the northern and the western periphery of the anticyclone is particularly strengthened (Fig.
14 7-8, Annex Figs VII-IX). Similar features are also observed at the higher pressure levels (700hPa, 500 hPa,
15 Annex Figs XI) as it was also the case for the April episode.

16 The IASI measurements (composite charts) at 3 km during the May 7-9, 2008 episode is shown in Fig. 7, while
17 for the lower and the upper troposphere (3 and 10km respectively) for the previous 2, 3 and 5 days are shown in
18 Annex Fig. X. During the May episode a high ozone area is observed in the lower and upper troposphere over
19 the N. Atlantic, which is more extended during the days preceding the episode peak.

20 As observed (Fig. 7, Annex Fig. X) the days before the episode and over the region covered by the anticyclone,
21 the concentrations of ozone in the upper troposphere (at 10 km) decrease, while the IASI measurements in the
22 lower troposphere (at 3 km) over central Europe show a small but geographically extended increase. It has to be
23 reminded that this phenomenon takes place exactly over the area where intense subsidence and low humidity
24 conditions have been detected, which could be considered as a direct independent evidence of the influence of
25 ozone concentrations from the upper troposphere, as it has been described and discussed in the previous
26 paragraphs. As also seen in Figure 7, over E. Europe and Russia a significant tropospheric ozone accumulation
27 occurs while a prevailing easterly flow from these high ozone areas is moving towards central and W. Europe. It
28 should be reminded that over central Europe extended strong subsidence associated with dry conditions and a
29 strong gradient of temperature anomalies are observed at the same time (Fig. 7, Annex Figs VII-X).

30 The above features are more clearly shown in Fig. 9 where the daily IASI satellite ozone measurements at 3 and
31 10 km level for the May 2008 episode are presented in a higher concentration resolution as well as in a more
32 restricted geographical domain (same as the CHIMERE model outputs) while the corresponding daily
33 meteorological charts of columnar precipitable water anomaly and geopotential heights at 500hPa and 700hPa
34 are shown in Annex Fig. XI. In fact, an extended region of negative columnar precipitable water anomalies (dry
35 conditions throughout the troposphere, indicating subsidence), prevailing over a vast area covering eastern and
36 central Europe is observed. As also seen in the April episode, this feature appears at the interface areas of the
37 high and low pressure systems while the same synoptic patterns occur at all examined tropospheric levels. In
38 addition, in Fig. 9 the graphical representation on the map of the daily evolution of the hourly surface ozone

1 concentrations at 15:00 h as recorded in the air pollution stations of the EEA-AirBase network (EEA, 2015)
2 during the 7-9 May period shows very high concentrations, exceeding 70 ppb, appearing simultaneously at many
3 locations from N. Italy to the British islands.

4 The results of the CHIMERE simulations for the May episode are shown in Fig. 10 and Annex Fig. XII, where
5 simulations of upper tropospheric tracer concentrations, simulations of photochemical production and
6 simulations of the ozone field (together with the iso-contours of the high tropospheric tracer) are presented. As
7 described earlier (cf Chapter 2), we use one tracer initialized within the model top layer in the upper troposphere
8 at about 11 km height. Inspecting qualitatively simulated concentrations at 3 km and 1.5 km altitude (Fig. 10,
9 Annex Fig. XII), it appears that the upper tropospheric tracer is clearly detected at these levels, indicating
10 downward transport from the upper troposphere to the boundary layer. Also in Fig. 10, the CHIMERE
11 simulations of the ozone fields at 3 km and 1.5 km altitudes (corresponding to 700hPa and 850hPa pressure
12 levels respectively) show high ozone concentrations over C. Europe, Italy and the W. Mediterranean. More
13 precisely, the highest upper tropospheric tracer influence occurs to the south of the N. European anticyclone and
14 it is progressively strengthened between 7 and 9 of May while high ozone is observed over the whole
15 anticyclonic area. In general, over that area the observed high values of the upper tropospheric tracer indicate
16 considerable subsidence at 5 km altitude (not shown), which becomes even stronger and more extended at the at
17 3 km and 1.5 km altitudes (Fig. 10). Therefore, the downward transport of upper tropospheric ozone is
18 influencing the lower troposphere and the boundary layer over the examined location. The observed patterns,
19 based on CHIMERE tracer simulations are quite consistent with the above described analysis based on
20 meteorological charts and IASI satellite measurements (Figs 7, 9). Overall, there is a good agreement between
21 the CHIMERE tracer simulations, the composite meteorological maps (Fig. 7) and the IASI satellite
22 measurements (Figs 7, 9), showing an extended region of subsiding dry air masses over eastern and central
23 Europe. As mentioned previously, during the afternoon hours the influence of the free troposphere on the
24 boundary layer is maximized through vertical mixing and thus the surface ozone concentrations could be
25 increased by the entrainment of subsiding ozone-rich air.

26 Regarding the comparison of the CHIMERE ozone field at 3 km with IASI at 700hPa there is in general a good
27 agreement over Italy and the central Mediterranean but there are differences in E. Europe (lower CHIMERE
28 values) and N. Europe (higher CHIMERE values), which might imply an underestimation of tropospheric
29 transport in E. Europe or overestimation of photochemistry in N. Europe. It should be reminded that similar
30 features appear also in the April episode, described previously. Nevertheless, as mentioned also for the April
31 episode, it should be kept in mind that for a direct quantitative comparison between CHIMERE and IASI data,
32 an application of IASI averaging kernels (smoothing functions) to CHIMERE output would be required
33 (Eremenko et al., 2008).

34 In Figure 11 the HYSPLIT back-trajectories during the episode at EMEP stations in Italy (IT04) and France
35 (FR10, FR14) show subsiding air masses arriving either from the north after performing a circular clockwise
36 movement or from the NE (E. Europe and Russia) where high tropospheric ozone concentrations have been
37 recorded by IASI (Figs. 7, 9). This is an additional confirmation that air masses originating from areas with high
38 tropospheric ozone concentrations transport ozone down to the ground, leading to high surface ozone values.

3.3.1 Ozone vertical profiles over Payerne, Uccle and Hohenpeissenberg during the May 2008 ozone episode

Due to the more complex nature of the May 7-9, 2008 ozone episode, the vertical ozone profiles at three European ozone sounding stations (Payerne-Switzerland, Hohenpeissenberg-Germany and Uccle-Belgium) were taken into account for the analysis and the respective vertical ozone measurements for each station are shown in Fig 12.

It has to be reminded that the Payerne site is one of the two Swiss stations, where the rural afternoon surface ozone values during the May episode were about 75 ppb (Fig 1). The composite meteorological maps (Figs 7, 8) indicate that the Payerne site was within the area influenced by subsidence. As observed in Fig. 12 a layer with the tropospheric ozone maximum concentration and low relative humidity is at the beginning of the event (on May 5) located between 5 and 6 km altitude. On May 7 a similar layer is seen at a somewhat lower altitude and on May 9 the tropospheric ozone maximum is found below 2000 m altitude. It has to be added that on May 12, at the end of the episode, the vertical ozone profile has changed completely and the ozone concentrations up to 6 km were about 60 ppb (not shown). As mentioned in the meteorological analysis of the episode (Figs 7, 8), the downward ozone transport from the ozone-rich lower troposphere to the boundary layer during the 5-9 May period with strong and persistent subsidence, is reflected clearly in the ozone profiles over Payerne (Fig. 12). Based on the observed conditions the plausible explanation is that an “ozone fumigation” of the boundary layer occurred between 5-9 May when the ozone levels increased by at least 20 ppb, following corresponding changes in the lower troposphere and indicating that this ozone event is related to downward transport of ozone from higher altitudes towards the boundary layer.

Similar observations could be made at Hohenpeissenberg and Uccle during the May 5-9 period (Fig. 12) where high ozone layers are also observed in the lower troposphere (around 80–100 ppb at 3-5 km on May 5 over both sites) and which clearly move downwards. The observed pattern is in agreement with the strong and persistent subsidence observed over the area during the examined days, as it was the case for Payerne. As shown in the vertical ozone soundings (Fig. 12) the high ozone tongue in the troposphere is going down rapidly at a 1-2 km /day rate. At the same time over the examined area the positive vertical velocity omega observed are associated with dry air (Fig. 7), indicating subsidence from the upper troposphere all the way down to the surface.

It has to be noticed at this point that vertical ozone profiles over the airports of Frankfurt and London carried out in the framework of the MOZAIC project (Marenco et al., 1998; Thouret et al., 2006) during the examined period show almost exactly the same picture (not shown).

Taking into account all the above information regarding the May episode, the following remarks regarding the tropospheric ozone distribution could be made:

a). The IASI satellite measurements shows that over N. Atlantic and to the west of the N. European anticyclone (present at all tropospheric pressure levels up to 300hPa) there are high amounts of ozone in the upper and lower troposphere.

b). The anticyclone transports these air masses from the west to the east in a clockwise rotating movement.

1 c). Over E. Europe and at the southern part of the anticyclone the air masses show strong signs of subsidence
2 (downward vertical velocities, dry air), which is particularly enhanced in the vicinity of the strong low-pressure
3 system over eastern Europe.

4 d). A significant ozone decrease in the upper troposphere is observed over the region covered by the N.
5 European anticyclone followed by a slight but extended ozone increase in the lower troposphere, located over the
6 area with intense subsidence (Central Europe) to the south of the anticyclone, with a several day time-shift.

7 In summarizing the observations on the May 7-9, 2008 episode an important question is to which extent the high
8 surface ozone levels observed at first in countries surrounding the western Mediterranean basin but also over a
9 large area in central Europe are influenced by entrainment of ozone rich air from higher layers of the
10 troposphere. An interesting feature is that during the examined period an extended area of strong and persistent
11 downward movement of air masses is observed over Central Europe and lasts for many days. Another important
12 observation is that the wind makes a circular downward motion around the anticyclone while this air current is
13 influenced from regions of very high tropospheric ozone surrounding the anticyclone (located over the observed
14 low pressure systems), according to IASI observations. The high ozone areas coincide very well with extended
15 areas of low humidity as observed in the 850hPa level charts but also at higher tropospheric levels. At the same
16 time, the vertical ozone profiles over Payerne, Uccle and Hohenpeissenberg show concentrations at 60-80 ppb on
17 the top of the boundary layer. It should be noticed that all the above sites are located more or less within the
18 region of high subsidence as the meteorological analysis shows. The CHIMERE tracer simulation experiments
19 are in very good agreement with the above observational analysis. As observed, during this episode the high
20 measured tropospheric ozone background values contribute very significantly, through subsidence, to enhanced
21 ozone values especially over France, Switzerland, northern Italy, and the Western Mediterranean basin, while
22 regional ozone photochemical production is the dominant factor for ozone enhancement over the British Islands,
23 the North Sea as well as on the Iberia peninsula and parts of France (Fig. 10, Annex Fig. XII).

24 Summarizing all the above, during the examined May episode as well as in the previous April episode, high
25 atmospheric pressures prevail over the Western Mediterranean and Central Europe while low-pressure systems
26 are observed in Eastern Europe. In general, both examined episodes (26-27 April 2008 and 7-9 May 2008) seem
27 to show comparable characteristics: At first, very strong positive omega anomalies (strong downward air motion
28 or subsidence) are observed over large areas including the Italian peninsula and Central Europe. At the same
29 time, strong negative specific humidity anomalies and negative precipitable water anomalies (indicating dry air
30 in the troposphere) are detected. An interesting feature is that the subsidence area is located at the interface of
31 positive and negative anomalies of geopotential heights and temperature. For the May episode, although the
32 major anticyclone seems to be located away from the Mediterranean basin to the north, a large-scale downward
33 transport towards the Mediterranean according to the meteorological analysis is observed. It comes out from the
34 above analysis that the interpretation of the back trajectory information is much more efficient if it is done in
35 combination with the examination of the meteorological charts (and ideally supported by satellite measurements
36 and modelling simulations), so that the regions with strong subsidence and dry air, are detected and taken into
37 account in the interpretation.

38

3.4 Diurnal variation of surface ozone and humidity during the ozone episodes

The measurements of surface ozone, relative humidity and temperature at the EMEP site at Ispra, Italy, (Jensen, 2016) have been used during the April and May episodes in order to observe the relationship between ozone and absolute humidity (Fig. 13), which could be a good indicator for the detection of the free tropospheric influence to the boundary layer being insensitive to the diurnal temperature variation. Absolute humidity is shown in units of hPa water vapor partial pressure, calculated from the relative humidity using the August-Roche-Magnus formula to find the saturated water vapor partial pressure at the measured temperature. The ozone concentrations show a characteristic diurnal variation with a minimum during the night, followed by a rapid increase in the morning when the nocturnal boundary layer breaks down and air from the above layer reaches the ground. For all days with high ozone during the May episode and for some days during the April episode the ozone increase at mid-day with the maximum vertical mixing is clearly anticorrelated with absolute humidity. This is compatible with the fact that the Ispra site, according to the meteorological analysis presented above, during the whole duration of the May episode is expected to be influenced by subsidence of dry air while during the April episode much stronger anticorrelations and lower absolute humidity levels are observed for some days. Based on the presented example, a first identification of possible direct tropospheric influence to the boundary layer air could be performed at air pollution monitoring stations potentially influenced by tropospheric subsidence.

4 Conclusions

In this paper, the investigation of the regional ozone episodes in the western Mediterranean and the surrounding countries is focused on the spring season.

Based on the ozone measurements at the EMEP rural ozone stations surrounding the western Mediterranean basin (as well as the EEA-AirBase network), it is observed that episodic periods of high ozone may last for several days and they can be detected at several countries at the same time.

An interesting result was that the examined ozone episodes are linked to meteorological conditions very similar to those observed in the eastern Mediterranean (Kalabokas et al., 2013; Doche et al., 2014; Kalabokas et al., 2015), related essentially with transport of ozone-rich tropospheric air masses and the atmospheric subsidence phenomenon. Based on the analysis of the selected springtime ozone episodes, the characteristics associated with high ozone concentrations close to the surface through important subsidence and in connection with high tropospheric ozone levels occurring in the wider area, are the following:

The geographic areas with observed deep tropospheric subsidence seem to be the transition regions between a high pressure system, located in the west sector, and a low pressure system located in the east sector, as shown in the corresponding charts of the geopotential heights. Over these areas, strong gradients of geopotential height and temperature are observed together with high omega vertical velocity values and low specific humidity values at the 850hPa as well as at higher tropospheric pressure levels. In addition, over the areas of deep tropospheric subsidence, negative temperature anomalies are observed at these levels and also high vector wind speeds, which means that subsidence is associated with strong advection. The above observational analysis is in very good agreement with IASI satellite measurements and CHIMERE tracer simulation experiments.

1 The present approach, using meteorological charts, IASI tropospheric ozone satellite measurements, CHIMERE
2 tracer simulations and back trajectories for the analysis of selected spring ozone episodes, shows to be quite
3 efficient in the analysis of atmospheric conditions and transport patterns associated with the episodes, which
4 need to be adequately described in numerical chemical-transport models used for simulations of air pollution. It
5 can be useful for the study of the tropospheric influence on the boundary layer and the ground surface, especially
6 for tracing large scale deep subsidence events (downward movements of generally dry air masses) when
7 analyzing surface (and vertical, if available) measurements at sites in the Mediterranean basin, where this
8 phenomenon is quite frequent, and also in other places worldwide. For example, similar field observations on the
9 influence of upper atmospheric layers on the boundary layer and surface measurements have been reported at
10 some locations in the United States (Parrish et al., 2010; Cooper et al., 2011; Langford et al., 2015). In addition,
11 the consideration for the analysis of ozone measurements presented above is in agreement with recent
12 observations in Atlantic and European regions indicating significant tropospheric (and even stratospheric)
13 influence to the surface and boundary layer ozone measurements (Trickl et al., 2010; Trickl et al., 2011; Logan
14 et al., 2012; Cuevas et al., 2013; Hess and Zbinden, 2013; Cooper et al., 2014; Parrish et al., 2014).

15 Regarding the environmental policy issues, it has to be underlined that for ozone, which is a pollutant regulated
16 by the EU, it appears that there are some time periods during the warm period of the year, lasting for several
17 days, when the free troposphere influences significantly the boundary layer to such extent that the air quality
18 standards might be exceeded. This phenomenon seems to be associated with an important impact of
19 photochemical ozone production following primary air pollutant emission at larger geographical scales,
20 including transport of air pollutants on a hemispheric scale from e.g. the US and China. The origin of the
21 atmospheric ozone entering the boundary layer might be upper tropospheric or stratospheric but it could also be
22 from the lower or the middle troposphere during stagnant regional conditions when photochemical ozone is
23 produced over the continent.

24 Further detailed and quantitative studies of this phenomenon appear to be needed to improve our understanding
25 of the mechanisms associated with intercontinental pollution as well as regional photochemical pollution could
26 be improved, already highlighted by the work of the HTAP (Hemispheric Transport of Air Pollution) project.
27 The Mediterranean region which seems to be mostly affected is the Eastern Mediterranean, where the 60 ppb EU
28 standard is very frequently exceeded during the warm period of the year and especially during July-August when
29 the atmospheric subsidence is a quite common atmospheric feature and almost quasi-permanent, but as shown in
30 the present study, also in the Western Mediterranean a similar mechanism could be observed, especially during
31 the spring season. The strategies for obtaining compliance with the EU ozone standard may need to be
32 reconsidered by taking into account the contribution from regional and intercontinental transport.

33

34 **Acknowledgements**

35 **Acknowledgement is made for the use of ECMWF's computing and archive facilities in this research.**

36 Acknowledgement is also made for the composite weather maps were provided by the NOAA/ESRL Physical
37 Sciences Division, Boulder Colorado from their Web site at <http://www.cdc.noaa.gov/>. The authors also
38 acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and

1 dispersion model and/or READY website (<http://www.ready.noaa.gov>) used in this publication. Also, Dr. A.
2 Volz-Thomas, FZ-Juelich, is acknowledged for interesting discussion as well as Dr. C. Repapis, Academy of
3 Athens, for useful comments on the manuscript. JRC is acknowledged for an EU grant (cat. 40) to one of the
4 authors (P. K.). Our particular thanks go to the operators of the following air quality monitoring stations: Cabo
5 de Creus in Spain, Morvan and Montandon in France, Payerne and Chaumont in Switzerland, Montelibretti and
6 Ispra in Italy and Giordan Lighthouse in Malta. This work was performed using HPC resources from GENCI-
7 TGCC (Grant 2016-[t20155017232]).

10 References

11 Beekmann, M. and Vautard R. (2010), A modelling study of photochemical regimes over Europe: robustness
12 and variability, *Atmos. Chem. Phys.*, 10, 10067-10084.

13 Beekmann, M., Ancellet, G. and Megie, G. 1994. Climatology of tropospheric ozone in southern Europe and its
14 relation to potential vorticity, *J. Geoph. Res.*, 99, 12841-12853.

15 Bonasoni, P., Stohl, A., Cristofanelli, P., Calzolari, F., Colombo, T. and Evangelisti, F. 2000. Background ozone
16 variations at Mt. Cimone station. *Atmos. Environ.* 34, 5183-5189.

17 Boynard, A., Clerbaux, C., Coheur, P.-F., Hurtmans, D., Turquety, S., George, M., Hadji-Lazaro, J., Keim, C.,
18 and Meyer-Arneke, J. 2009. Measurements of total and tropospheric ozone from IASI: comparison with
19 correlative satellite, ground-based and ozonesonde observations, *Atmos. Chem. Phys.*, 9, 6255–6271,
20 doi:10.5194/acp-9-6255-2009.

21 Clarisse, L., R'Honi, Y., Coheur, P.-F., Hurtmans, D., and Clerbaux, C., 2011. Thermal infrared nadir
22 observations of 24 atmospheric gases, *Geophys. Res. Lett.*, 38, L10802, doi:10.1029/2011GL047271.

23 Clerbaux, C., Boynard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D., Pommier, M.,
24 Razavi, A., Turquety, S., Wespes, C., and Coheur, P.-F. 2009. Monitoring of atmospheric composition using the
25 thermal infrared IASI/MetOp sounder, *Atmos. Chem. Phys.*, 9, 6041–6054, doi:10.5194/acp-9-6041-2009.

26 Coheur, P.-F., Barret, B., Turquety, S., Hurtmans, D., Hadji-Lazaro, J., and Clerbaux, C. 2005. Retrieval and
27 characterization of ozone vertical profiles from a thermal infrared nadir sounder, *J. Geophys. Res.*, 110, D24303,
28 doi:10.1029/2005JD005845.

29 Cooper, O.R., Oltmans, S.J., Johnson, B.J., Brioude, J., Angevine, W., Trainer, M., Parrish, D. D., Ryerson, T.
30 R., Pollack, I., Cullis, P. D., Ives, M.,A., Tarasick D.W., Al-Saadi, J., Stajner, I. 2011. Measurement of western
31 U.S. baseline ozone from the surface to the tropopause and assessment of downwind impact regions. *J. Geophys.*
32 *Res* 116:D00V03. doi:10.1029/2011JD016095

33 Cooper, O. R., Gao, R.-S., Tarasick, D., Leblanc, T., Sweeney, C. 2012. Long-term ozone trends at rural ozone
34 monitoring sites across the United States, 1990–2010. *J. Geophys. Res* 117: D22307.
35 doi:10.1029/2012JD018261

1 Cooper, O. R., Parrish, D. D., Ziemke, J., Balashov, N. V., Cupeiro, M., Galbally, I. E., Gilge, S., Horowitz,
2 L., Jensen, N. R., Lamarque, J.-F., Naik, V., Oltmans, S. J., Schwab, J., Shindell, D. T., Thompson, A. M.,
3 Thouret, V., Wang, Y., Zbinden, R. M. 2014. Global distribution and trends of tropospheric ozone: An
4 observation-based review. *Elementa: Sci. of the Anthropocene*. 2: 000029, doi:
5 10.12952/journal.elementa.000029

6 Coman, A., Foret, G., Beekmann, M., Eremenko, M., Dufour, G., Gaubert, B., Ung, A., Schmechtig, C., Flaud,
7 J.-M., and Bergametti, G. 2012. Assimilation of IASI partial tropospheric columns with an Ensemble Kalman
8 Filter over Europe, *Atmos. Chem. Phys.*, 12, 2513–2532, doi:10.5194/acp-12-2513-2012.

9 Cristofanelli, P., Scheel, H.-E., Steinbacher, M., Saliba, M., Azzopardi, F., Ellul, R., Froehlich, M., Tositti, L.,
10 Brattich, E., Maione, M., Calzolari, F., Duchi, R., Landi, T.C., Marinoni, A. and Bonasoni P. 2015. Long-term
11 surface ozone variability at Mt. Cimone WMO/GAW global station (2165 m a.s.l., Italy). *Atmos. Environ.* 101,
12 23-33.

13 Cuevas, E., González, Y., Rodríguez, S., Guerra, J.C., Gómez-Peláez, A.J., Alonso Pérez, S., Bustos, J., Milford,
14 C. 2013. Assessment of atmospheric processes driving ozone variations in the subtropical North Atlantic free
15 troposphere, *Atmos. Chem. Phys* 13:1973–1998.

16 Delmas, R., Mégie, G., and Peuch, V.-H. 2005. *Physique et chimie de l’atmosphère*, Ed. Belin, Coll. Echelles,
17 640

18 Doche, C., Dufour, G., Foret, G., Eremenko, M., Cuesta, J., Beekmann, M., and Kalabokas, P. 2014.
19 Summertime tropospheric-ozone variability over the Mediterranean basin observed with IASI, *Atmos. Chem.*
20 *Phys.*, 14, 10589–10600.

21 Draxler, R.R. and Rolph, G.D. 2015. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory)
22 Model access via NOAA ARL READY Website (<http://www.arl.noaa.gov/HYSPLIT.php>). NOAA Air
23 Resources Laboratory, College Park, MD.

24 EEA, 2015: <http://www.eea.europa.eu/themes/air/air-quality/map/airbase>

25 **EMEP, 2015. EMEP, Chemical Coordinated Center, www.emep.int**

26 Dufour, G., Eremenko, M., Orphal, J., and Flaud, J.-M. 2010. IASI observations of seasonal and day-to-day
27 variations of tropospheric ozone over three highly populated areas of China: Beijing, Shanghai, and Hong Kong,
28 *Atmos. Chem. Phys.*, 10, 3787–3801, doi:10.5194/acp-10-3787-2010.

29 Dufour, G., Eremenko, M., Griesfeller, A., Barret, B., LeFlochmoën, E., Clerbaux, C., Hadji-Lazaro, J., Coheur,
30 P.-F. and Hurtmans D., Validation of three scientific ozone products retrieved from IASI spectra using
31 ozonesondes, *Atmos. Meas. Tech.*, 5, 611-630, 2012.

32 Dufour, G., Eremenko, M., Cuesta, J., Doche, C., Foret, G., Beekmann, M., Cheiney, A., Wang, Y., Cai, Z., Liu,
33 Y., Takigawa, M., Kanaya, Y., and Flaud, J.-M.: Springtime daily variations in lower-tropospheric ozone over
34 east Asia: the role of cyclonic activity and pollution as observed from space with IASI, *Atmos. Chem. Phys.*, 15,
35 10839-10856, doi:10.5194/acp-15-10839-2015, 2015.

1 Eremenko, M., Dufour, G., Foret, G., Keim, C., Orphal, J., Beekmann, M., Bergametti, G., and Flaud, J.-M.
2 2008. Tropospheric ozone distributions over Europe during the heat wave in July 2007 observed from infrared
3 nadir spectra recorded by IASI, *Geophys. Res. Lett.*, 35, L18805, doi:10.1029/2008GL034803.

4 Fishman, J., Wozniak, A. E., and Creilson, J. K. 2003. Global distribution of tropospheric ozone from satellite
5 measurements using the empirically corrected tropospheric ozone residual technique: Identification of the
6 regional aspects of air pollution, *Atmos. Chem. Phys.*, 3, 893–907, doi:10.5194/acp-3-893-2003.

7 Foret, G., Hamaoui, L., Schmechtig, C., Eremenko, M., Keim, C., Dufour, G., Boynard, A., Coman, A., Ung, A.,
8 and Beekmann, M. 2009. Evaluating the potential of IASI ozone observations to constrain simulated surface
9 ozone concentrations, *Atmos. Chem. Phys.*, 9, 8479–8491, doi:10.5194/acp-9-8479-2009.

10 Fuhrer, J. 2009. Ozone risk for crops and pastures in present and future climates, *Naturwissenschaften*, 96, 173–
11 194.

12 Hess, P.G., Zbinden, R., 2013. Stratospheric impact on tropospheric ozone variability and trends: 1990–2009,
13 *Atmos. Chem. Phys.*, 13, 649–674.

14 George, M., Clerbaux, C., Hurtmans, D., Turquety, S., Coheur, P.-F., Pommier, M., Hadji-Lazaro, J., Edwards,
15 D. P., Worden, H., Luo, M., Rinsland, C., and McMillan, W. 2009. Carbon monoxide distributions from the
16 IASI/METOP mission: evaluation with other space-borne remote sensors, *Atmos. Chem. Phys.*, 9, 8317–8330,
17 doi:10.5194/acp-9-8317-2009.

18 Gerasopoulos, E., Kouvarakis, G., Vrekoussis, M., Kanakidou, M., Mihalopoulos, N., 2005. Ozone variability in
19 the marine boundary layer of the Eastern Mediterranean based on 7-year
20 Observations, *J. Geoph. Res.*, 110, D15309.

21 Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C. 2006. Estimates of global
22 terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmos.*
23 *Chem. Phys.*, 6, 3181–3210, www.atmos-chem-phys.net/6/3181/2006/

24 HTAP report, 2010. Hemispheric Transport of Air Pollution 2010. Part A: Ozone and Particulate Matter, Air
25 Pollution Studies No 17, United Nations Economic Commission for Europe, ISBN 978-92-1-117043-6

26 IPCC: Climate Change 2007: The Physical Science Basis, Contribution of Working Group I to the Fourth
27 Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D.,
28 Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press,
29 Cambridge, United Kingdom and New York, NY, USA, 2007.

30 Johnson, J. E., Sundet, J. K., and Tarrason, L. 2001. Model calculations of present and future levels of ozone and
31 ozone precursors with a global and regional model, *Atmos. Environ.*, 35, 525–537.

32 Jensen, N.R. (2016), personal communication.

33 Kalabokas, P. D., Viras, L. G., Bartzis, J. G., and Repapis, C. C. 2000. Mediterranean rural ozone characteristics
34 around the urban area of Athens, *Atmosph. Environ.*, 34, 5199–5208.

1 Kalabokas, P. D. and Repapis, C. C. 2004. A climatological study of rural surface ozone in central Greece,
2 *Atmos. Chem. Phys.*, 4, 1139–1147, doi:10.5194/acp-4-1139-2004.

3 Kalabokas, P. D., Volz-Thomas, A., Brioude, J., Thouret, V., Cammas, J.-P., and Repapis, C. C. 2007. Vertical
4 ozone measurements in the troposphere over the Eastern Mediterranean and comparison with Central Europe,
5 *Atmos. Chem. Phys.*, 7, 3783–3790, doi:10.5194/acp-7-3783-2007.

6 Kalabokas, P. D., Mihalopoulos, N., Ellul, R., Kleanthous, S., and Repapis, C. C. 2008. An investigation of the
7 meteorological and photochemical factors influencing the background rural and marine surface ozone levels in
8 the Central and Eastern Mediterranean, *Atmos. Environ.*, 42, 7894–7906

9 Kalabokas, P. D., Cammas, J.-P., Thouret, V., Volz-Thomas, A., Boulanger, D., and Repapis, C. C. 2013.
10 Examination of the atmospheric conditions associated with high and low summer ozone levels in the lower
11 troposphere over the eastern Mediterranean, *Atmos. Chem. Phys.*, 13, 10339–10352, doi:10.5194/acp-13-10339-
12 2013.

13 Kalabokas, P. D., Thouret, V., Cammas, J.-P., Volz-Thomas, A., Boulanger, D., and Repapis, C. C. 2015. The
14 geographical distribution of meteorological parameters associated with high and low summer ozone levels in the
15 lower troposphere and the boundary layer over the Eastern Mediterranean (Cairo case). *Tellus B*, 67, 27853,
16 <http://dx.doi.org/10.3402/tellusb.v67.27853>

17 Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G.,
18 Woolen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C., Wang,
19 J., Leetmaa, A., Reynolds, R., Jenne, R., and Joseph, D. 1996. The NCEP/NCAR Reanalysis 40-year Project, *B.*
20 *Am. Meteorol. Soc.*, 77, 437–471.

21 Kleanthous, S., Vrekoussis, M., Mihalopoulos, N., Kalabokas, P. and Lelieveld, J. 2014. On the temporal and
22 spatial variation of ozone in Cyprus. *Sc. of the Tot. Environ.* 476–477 (2014), 677–687.

23 Kourtidis, K., Zerefos, C., Rapsomanikis, S., Simeonov, V., Balis, D., Perros, P. E., Thomson, A. M., Witte, J.,
24 Calpini, B., Sharobiem, W. M., Papayiannis, A., Mihalopoulos, N., and Drakou, R. 2002. Regional levels of
25 ozone in the troposphere over eastern Mediterranean, *J. Geophys. Res.*, 107, 8140, doi:10.1029/2000JD000140.

26 Kouvarakis, G., Vrekoussis, M., Mihalopoulos, N., Kourtidis, K., Rappenglueck, B., Gerasopoulos, E., and
27 Zerefos, C. 2002. Spatial and temporal variability of tropospheric ozone in the boundary layer above the Aegean
28 Sea (eastern Mediterranean), *J. Geophys. Res.*, 107, 8137, doi:10.1029/2000JD000081.

29 Kuenen, J. J. P., Visschedijk, A. J. H., Jozwicka, M. and Denier van der Gon, H. A. C. 2014. TNO-MACC_II
30 emission inventory; a multi-year (2003–2009) consistent high-resolution European emission inventory for air
31 quality modeling, *Atmos. Chem. Phys.*, 14, 10963–10976, doi:10.5194/acp-14-10963-2014, 2014.

32 Langford, A.O., Sen, C.J., Alvarez, R.J., Brioude, J., Cooper, O.R., Holloway, J.S., Lind, M.Y., Marchbanksa,
33 R.D., Pierce, R.B., Sandberg, S.P., Weickmann, A.M., Williams, E.J. 2015. An Overview of the 2013 Las
34 Vegas Ozone Study (LVOS): Impact of stratospheric intrusions and long-range transport on surface air quality.
35 *Atmos. Environ.*, 109, 305–322.

- 1 Levy, J. I., Carrothers, T. J., Tuomisto, J. T., Hammitt, J. K., and Evans, J. S. 2001. Assessing the Public Health
2 Benefits of Reduced Ozone Concentrations, *Environ. Health Perspect.*, 109, 1215– 1226.
- 3 Lelieveld, J., Berresheim, H., Borrmann, S., Crutzen, P. J., Dentener, F. J., Fischer, H., Feichter, J., Flatau, P. J.,
4 Heland, J., Holzinger, R., Korrmann, R., Lawrence, M. G., Levin, Z., Markowicz, K. M., Mihalopoulos, N.,
5 Minikin, A., Ramanathan, V., de Reus, M., Roelofs, G. J., Scheeren, H. A., Sciare, J., Schlager, H., Schultz, M.,
6 Siegmund, P., Steil, B., Stephanou, E. G., Stier, P., Traub, M., Warneke, C., Williams, J., and Ziereis, H. 2002.
7 Global air pollution crossroads over the Mediterranean, *Science*, 298, 794–799.
- 8 Lelieveld, J. 2009. Air pollution and climate, in: *The Physical Geography of the Mediterranean*, edited by:
9 Woodward, J. C., Oxford University Press, 599–614.
- 10 Li, Q., Jacob, D. J., Logan, J. A., Bey, I., Yantosca, R. M., Liu, H., Martin, R. V., Fiore, A. M., Field, B. D.,
11 Duncan, B. N., and Thouret, V. 2001. A tropospheric ozone maximum over the Middle East, *Geophys. Res.*
12 *Let.*, 28, 3235–3238, doi:10.1029/2001GL013134.
- 13 Liu, X., Chance, K. V., Sioris, C. E., Spurr, R. J. D., Kurosu, T. P., Martin, R. V., and Newchurch, M. J. 2005.
14 Ozone profile and tropospheric ozone retrievals from Global Ozone Monitoring Experiment: Algorithm
15 description and validation, *J. Geophys. Res.*, 110, D20307, doi:10.1029/2005JD006240.
- 16 Liu, J. J., Jones, D. B. A., Worden, J. R., Noone, D., Parrington, M., and Kar, J. 2009. Analysis of the
17 summertime build-up of tropospheric ozone abundances over the Middle East and North Africa as observed by
18 the Tropospheric Emission Spectrometer instrument, *J. Geophys. Res.*, 114, D05304,
19 doi:10.1029/2008JD010993.
- 20 Logan, J. A., Staehelin, J., Megretskaia, I. A., Cammas, J.-P., Thouret, V., Claude, H., De Backer, H.,
21 Steinbacher, M., Scheel, H.-E., Stübi, R., Fröhlich M. and Derwent R. 2012. Changes in ozone over Europe:
22 Analysis of ozone measurements from sondes, regular aircraft (MOZAIC) and alpine surface sites, *J. Geophys.*
23 *Res.*, 117, D09301, doi:10.1029/2011JD016952.
- 24 Marengo, A., Thouret, V., Nédélec, P., Smit, H., Helten, M., Kley, D., Karcher, F., Simon, P., Law, K., Pyle, J.,
25 Poschmann, G., Von Wrede, R., Hume, C., and Cook, T. 1998. Measurements of ozone and water vapor by
26 Airbus in-service aircraft: The MOZAIC airborne program. An overview, *J. Geophys. Res.*, 103, 25631–25642.
- 27 Menut, L., Bessagnet, B., Khvorostyanov, D., Beekmann, M., Blond, N., Colette, A., Coll, I., Curci, G., Foret,
28 G., Hodzic, A., Mailler, S., Meleux, F., Monge, J.-L., Pison, I., Siour, G., Turquety, S., Valari, M., Vautard, R.,
29 and Vivanco, M. G. 2013. CHIMERE 2013: a model for regional atmospheric composition modelling, *Geosci.*
30 *Model Dev.*, 6, 981–1028, doi:10.5194/gmd-6-981-2013.
- 31 Millán, M. M., Salvador, R., Mantilla, E., and Kallos, G. 1997. Photo-oxidant dynamics in the western
32 Mediterranean in summer: Results from European research projects, *J. Geophys. Res.* 102: 8811–23.
- 33 Millan, M.M., Mantilla, E., Salvador, R., Carratalá, R., Sanz, M. J., Alonso, L., Gangioti, G., and Navazo, M.
34 2000. Ozone cycles in the western Mediterranean basin: Interpretation of monitoring data in complex coastal
35 terrain. *J. Appl. Meteor.* 39, 487– 508.

- 1 Monks, P. S., 2000. A review of the observations and origins of the spring ozone maximum, *Atmos. Environ.*,
2 34, 3545–3561.
- 3 Monks, P. S., Archibald, A. T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law,
4 K. S., Mills, G. E., Stevenson, D. S., Tarasova, O., Thouret, V., von Schneidemesser E., Sommariva, R., Wild,
5 O., and Williams, M. L. 2015. Tropospheric ozone and its precursors from the urban to the global scale from air
6 quality to short-lived climate forcer, *Atmos. Chem. and Phys.*, 15, 8889-8973.
- 7 Nolle, M., Ellul, R., Heinrich, G., and Güsten, H. 2002. A longterm study of background ozone concentrations in
8 the central Mediterranean—diurnal and seasonal variations on the island of Gozo, *Atmospheric Environment* 36:
9 1391–402.
- 10 Paoletti, E. 2006. Impact of ozone on Mediterranean forests: A Review, *Environ. Pollut.*, 144, 463–474.
- 11 Parrish, D. D., Aikin, K. C., Oltmans, S. J., Johnson, B. J., Ives, M., and Sweeny, C. 2010. Impact of transported
12 background ozone inflow on summertime air quality in a California ozone exceedance area, *Atmos. Chem.*
13 *Phys.*, 10, 10093–10109, doi:10.5194/acp-10-10093-2010.
- 14 Parrish, D.D., Law, K.S., Staehelin, J., Derwent, R., Cooper, O.R., Tanimoto, H., Volz-Thomas, A., Gilge, S.,
15 Scheel, H.-E., Steinbacher, M., and Chan, E. 2013. Lower tropospheric ozone at northern mid-latitudes:
16 Changing seasonal cycle. *Geophys. Res. Lett* 40: 1631–1636. doi:10.1002/grl.50303.
- 17 Parrish, D. D., Lamarque, J.-F., Naik, V., Horowitz, L., Shindell, D. T., Staehelin, J., Derwent, R., Cooper, O.
18 R., Tanimoto, H., Volz-Thomas, A., Gilge, S., Scheel, H.-E., Steinbacher, M., and Fröhlich, M. 2014. Long-term
19 changes in lower tropospheric baseline ozone concentrations: Comparing chemistry-climate models and
20 observations at northern midlatitudes, *J. Geophys. Res. Atmos.*, 119, 5719–5736, doi:10.1002/2013JD021435.
- 21 Querol X., Alastuey A., Orto A., Pallares M., Reina F., Dieguez JJ., Mantilla E., Escudero M., Alonso L.,
22 Gangoiti G., Millán M. 2016. On the origin of the highest ozone episodes in Spain. *Science of the Total*
23 *Environment*, 572, 379-389.
- 24 Richards, N. A. D., Arnold, S. R., Chipperfield, M. P., Miles, G., Rap, A., Siddans, R., Monks, S. A., and
25 Hollaway, M. J. 2013. The Mediterranean summertime ozone maximum: global emission sensitivities and
26 radiative impacts, *Atmos. Chem. Phys.*, 13, 2331–2345, doi:10.5194/acp-13-2331-2013.
- 27 Rodwell, M. J. and Hoskins, B. J. 1996. Monsoons and the dynamics of deserts, *Q. J. R. Meteorol. Soc.*, 122,
28 1385–1404, doi:10.1002/qj.49712253408.
- 29 Rodwell, M. J. and Hoskins, B. J. 2001. Subtropical anticyclones and summer monsoons, *J. Climate*, 14, 3192–
30 3211.
- 31 Rouil L., C.Honore, R.Vautard, M.Beekmann, B.Bessagnet, L.Malherbe, F.Meleux, A.Dufour, C.Elichegaray, J-
32 M.Flaud, L.Menuet, D.Martin, A.Peuch, V-H.Peuch, N.Poisson, 2009, PREV'AIR : an operational forecasting
33 and mapping system for air quality in Europe, *BAMS*, DOI: 10.1175/2008BAMS2390.1
- 34 Safieddine, S., Boynard, A., Coheur, P.-F., Hurtmans, D., Pfister, G., Quennehen, B., Thomas, J. L., Raut, J.-C.,
35 Law, K. S., Klimont, Z., Hadji-Lazaro, J., George, M., and Clerbaux C. 2014. Summertime tropospheric ozone

1 assessment over the Mediterranean region using the thermal infrared IASI/MetOp sounder and the WRF-Chem
2 model, *Atmos. Chem. Phys.*, 14, 10119–10131, doi:10.5194/acp-14-10119-2014.

3 Sánchez, M. L., García, M. A., Pérez, I. A., and de Torre, B. 2008. Evaluation of surface ozone measurements
4 during 2000–2005 at a rural area in the upper Spanish plateau, *J. Atmos. Chem.*, 60, 137–152.

5 Schürmann, G. J., Algieri, A., Hedgecock, I. M., Manna, G., Pirrone, N., and Sprovieri, F. 2009. Modeling local
6 and synoptic scale influences on ozone concentrations in a topographically complex region of Southern Italy,
7 *Atmos. Environ.*, 43, 4424–4434.

8 Seinfeld, J. H., Pandis, S. N., 2006. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*,
9 2nd Edition, Wiley.

10 Sicard, P., De Marco, A., Troussier, F., Renou, C., Vas, N., Paoletti, E. 2013. Decrease in surface ozone
11 concentrations at Mediterranean remote sites and increase in the cities. *Atmos. Environ* 79: 705–715.

12 Stohl, A., Eckhardt, S., Forster, C., James, P., and Spichtinger, N. 2002. On the pathways and timescales of
13 intercontinental air pollution transport, *J. of Geoph. Res(Atm.)*, 107(D23): 4684-4700.

14 Thouret, V., Cammas, J.-P., Sauvage, B., Athier, G., Zbinden, R., Nédélec, P., Simon, P., and Karcher, F. 2006.
15 Tropopause referenced ozone climatology and inter-annual variability (1994–2003) from the MOZAIC
16 programme, *Atmos. Chem. Phys.*, 6, 1033–1051, doi:10.5194/acp-6-1033-2006.

17 Trickl, T., Feldmann, H., Kanter, H.-J., Scheel, H.-E., Sprenger, M., Stohl, A., and Wernli, H., 2010. Forecasted
18 deep stratospheric intrusions over Central Europe: case studies and climatologies, *Atmos. Chem. Phys.*, 10, 499–
19 524, doi:10.5194/acp-10-499-2010.

20 Trickl, T., Bärtsch-Ritter, N., Eisele, H., Furger, M., Mücke, R., Sprenger, M., and Stohl, A. 2011. High-ozone
21 layers in the middle and upper troposphere above Central Europe: potential import from the stratosphere along
22 the subtropical jet stream, *Atmos. Chem. Phys.*, 11, 9343–9366, doi:10.5194/acp-11-9343-2011.

23 Tyrllis, E., Lelieveld, J., and Steil, B. 2013. The summer circulation in the eastern Mediterranean and the Middle
24 East: influence of the South Asian Monsoon, *Clim. Dynam.*, 40, 1103–1123, doi:10.1007/s00382-012-1528-4.

25 Tyrllis, E., Škerlak, B., Sprenger, M., Wernli, H., Zittis, G., and Lelieveld, J., 2014. On the linkage between the
26 Asian summer monsoon and tropopause fold activity over the eastern Mediterranean and the Middle East, *J.*
27 *Geophys. Res. Atmos.*, 119, 3202–3221, doi:10.1002/2013JD021113.

28 Velchev, K., Cavalli, F., Hjorth, J., Marmer, E., Vignati, E., Dentener, F., and Raes, F., 2011. Ozone over the
29 Western Mediterranean Sea – results from two years of shipborne measurements, *Atmos. Chem. Phys.*, 11, 675–
30 688, doi:10.5194/acp-11-675-2011.

31 Worden, H. M., Logan, J. A., Worden, J. R., Beer, R., Bowman, K., Clough, S. A., Eldering, A., Fisher, B. M.,
32 Gunson, M. R., Herman, R. L., Kulawik, S. S., Lampel, M. C., Luo, M., Megretskaja, I. A., Osterman, G. B., and
33 Shephard, M.W. 2007. Comparisons of Tropospheric Emission Spectrometer (TES) ozone profiles to ozone-
34 sondes: Methods and initial results, *J. Geophys. Res.*, 112, D03309, doi:10.1029/2006JD007258.

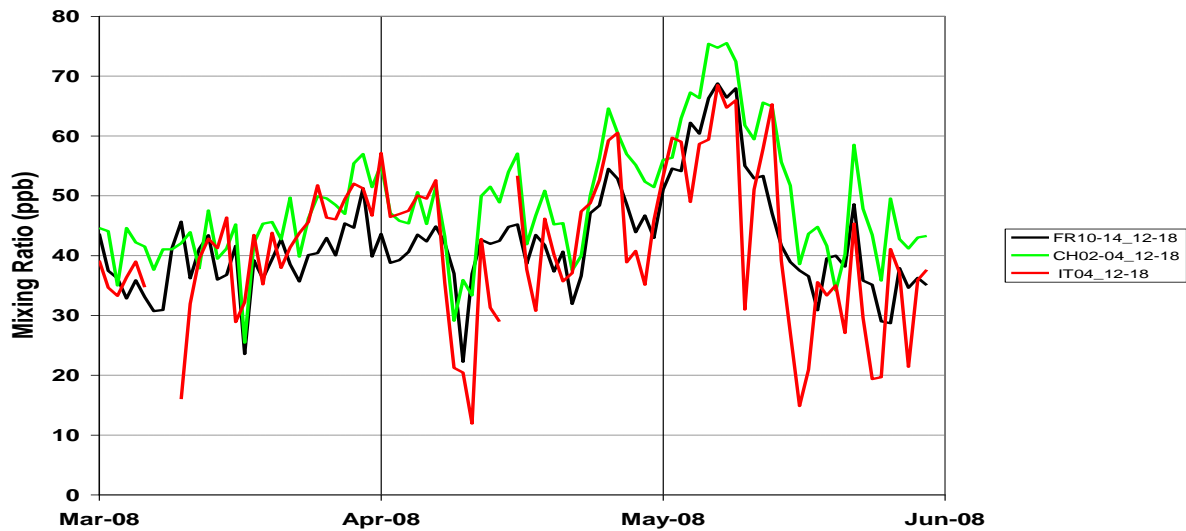
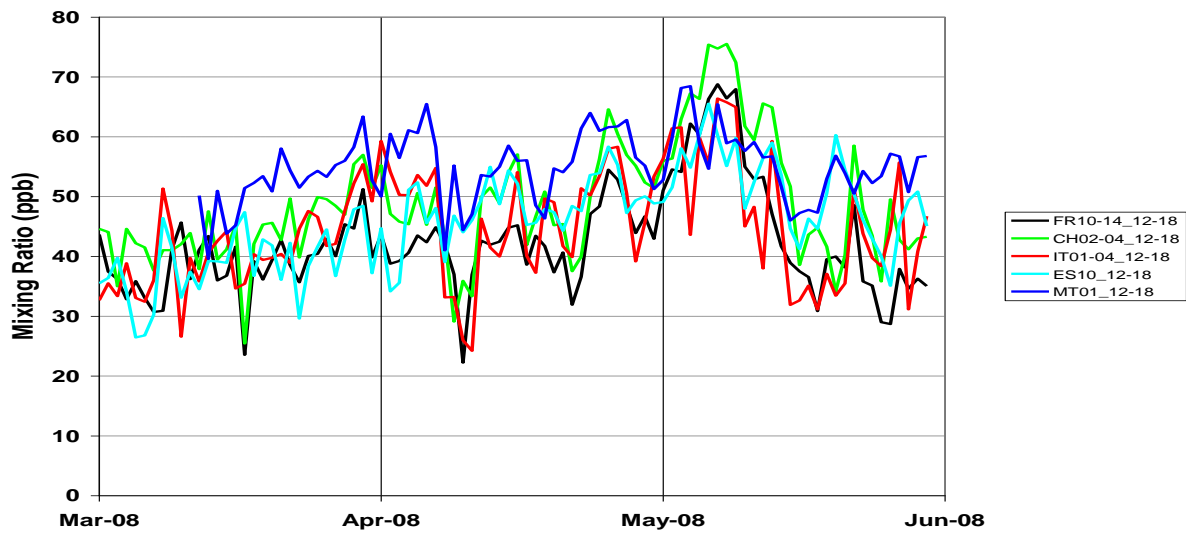
35 WOUDC, 2015: <http://woudc.org/data/explore.php>

1 Zanis, P., Hadjinicolaou, P., Pozzer, A., Tyrlis, E., Dafka, S., Mihalopoulos, N., and Lelieveld, J. 2014.
2 Summertime free-tropospheric ozone pool over the eastern Mediterranean/Middle East, *Atmos. Chem. Phys.*, 14,
3 115–132, doi:10.5194/acp-14-115-2014.

4

5

1

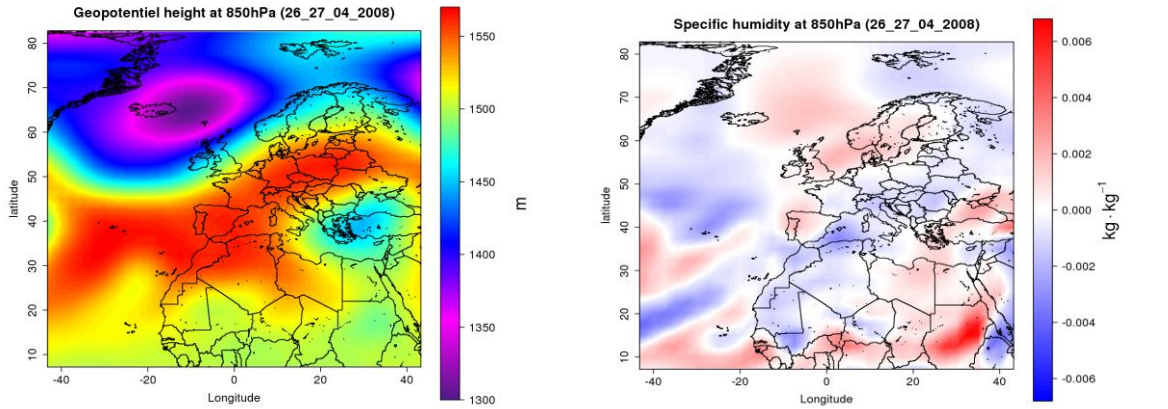


3

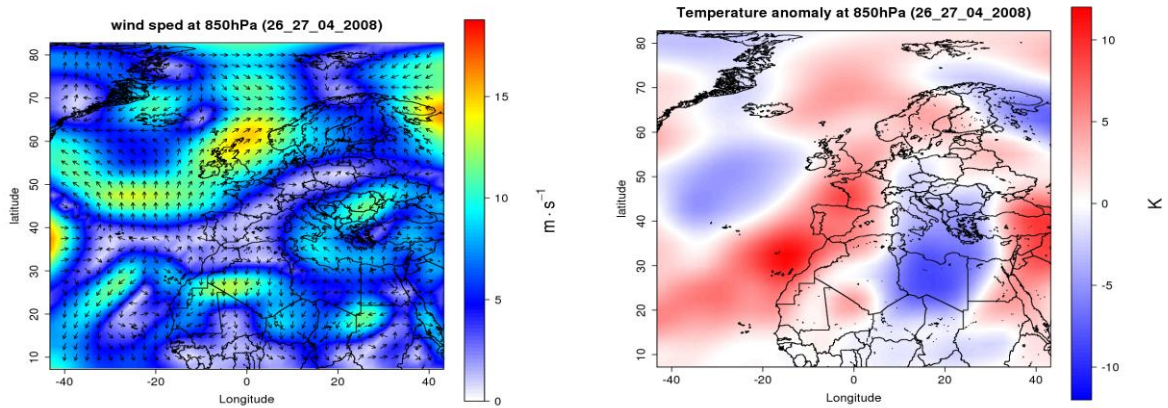
4 **Figure 1: (Upper panel): Day-to-day variation of the 12:00 - 18:00 afternoon ozone (in ppb) on the EMEP**
5 **stations surrounding the Western Mediterranean basin for spring 2008: Spain (ES10, light blue), France**
6 **(FR10-14, black), Switzerland (CH02-04, green), Italy (IT01-04, red), Malta (MT01, blue).**

7 **(Lower panel): Day-to-day variation of the 12:00 - 18:00 afternoon ozone (in ppb) on the EMEP stations**
8 **close to the Western Mediterranean basin and around the Alpine region for spring 2008: France (FR10-**
9 **14, black), Switzerland (CH02-04, green), Italy (IT04-JRC, red).**

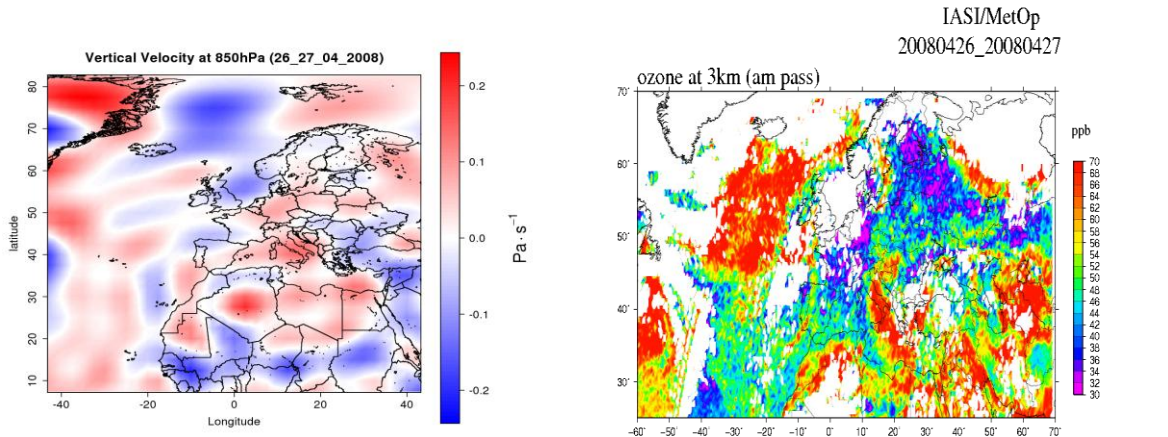
10



1



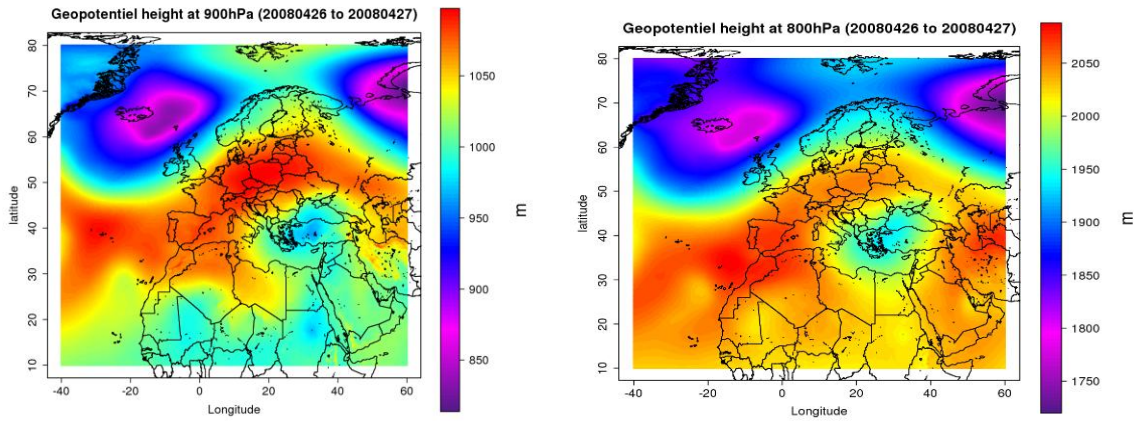
2



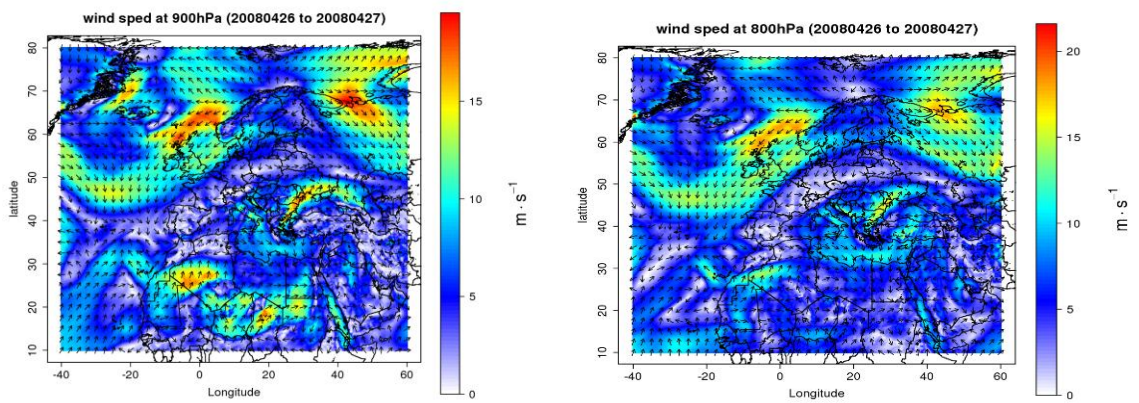
3

4 **Figure 2: Composite NOAA/ESRL weather maps of geopotential height (upper panel-left, in m), specific**
 5 **humidity anomaly (upper panel-right, in Kg/Kg), vector wind (middle panel-left, in m/s), air temperature**
 6 **anomaly (middle panel-right, in K), omega vertical velocity (lower panel-left, in Pa/s) and IASI satellite**
 7 **ozone concentrations at 3km (lower panel-right, in ppb) for the high ozone episode of 26-27 April 2008.**

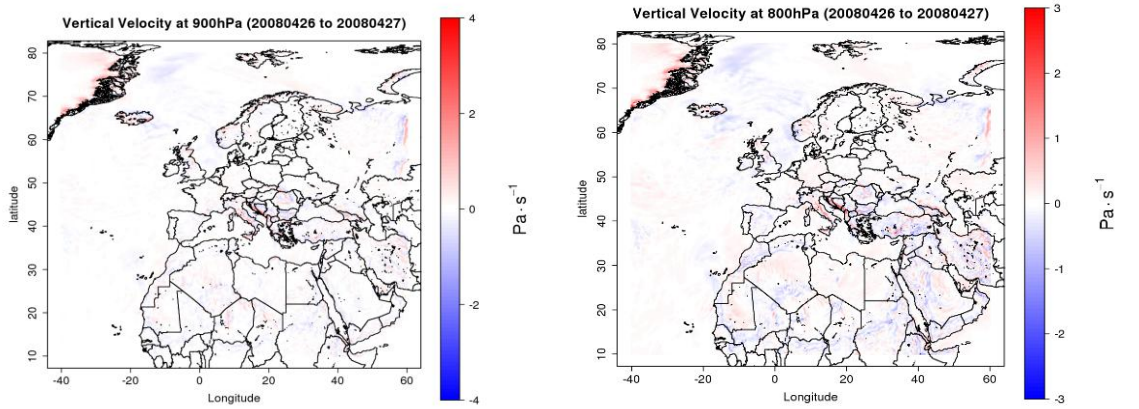
8



1



2

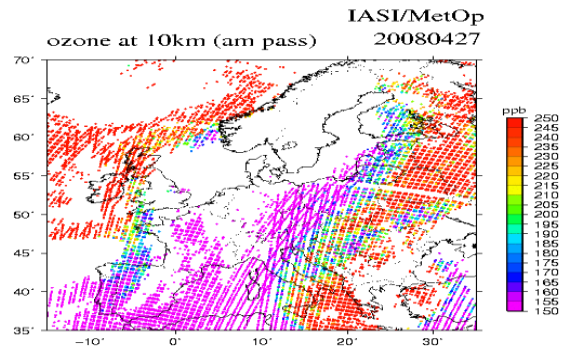
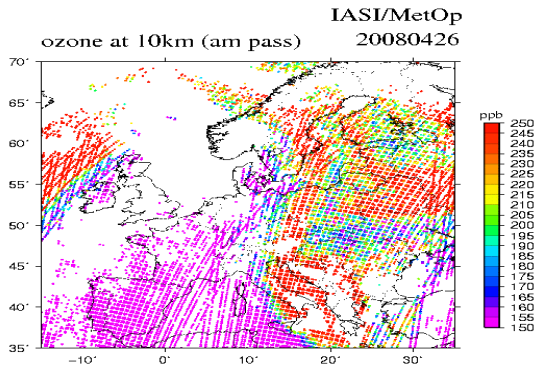


3

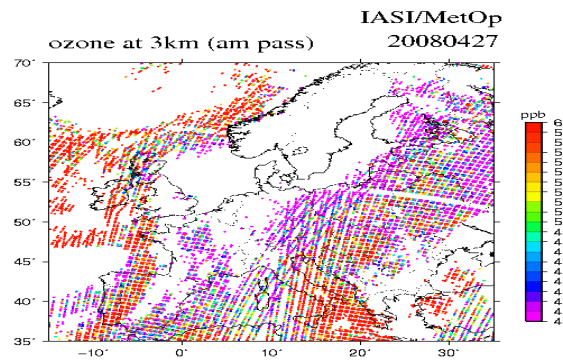
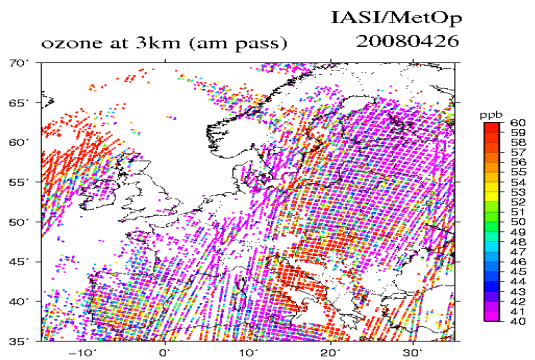
4

5 **Figure 3: Composite ECMWF meteorological charts of geopotential height (upper panel, in m), wind**
 6 **speed (middle panel, in m/s) and vertical velocity (lower panel, in Pa/s) at 900hPa (left) and 800 hPa**
 7 **(right) for the high ozone episode of 26-27 April 2008.**

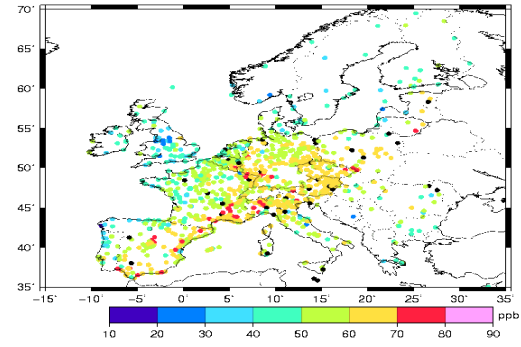
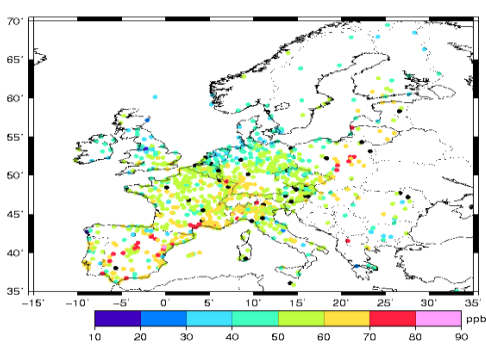
8



1



2



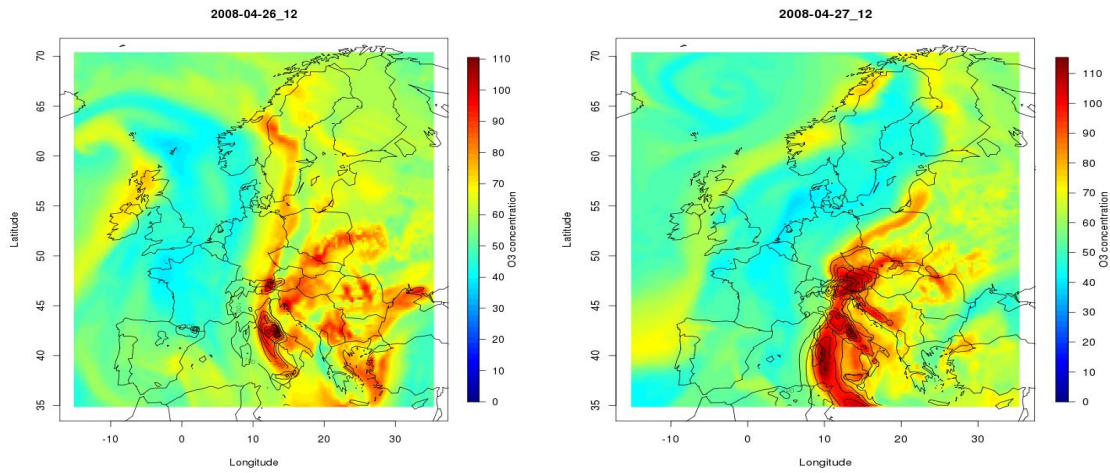
3

4 **Figure 4: Daily IASI satellite ozone measurements (ppb) at 10 km level (upper panel, in ppb) and at 3km**
 5 **level (middle panel, in ppb) for April 26, 2008 (left column) and April 27, 2008 (right column). Values**
 6 **outside the scale range are set up to the upper and lower color code respectively.**

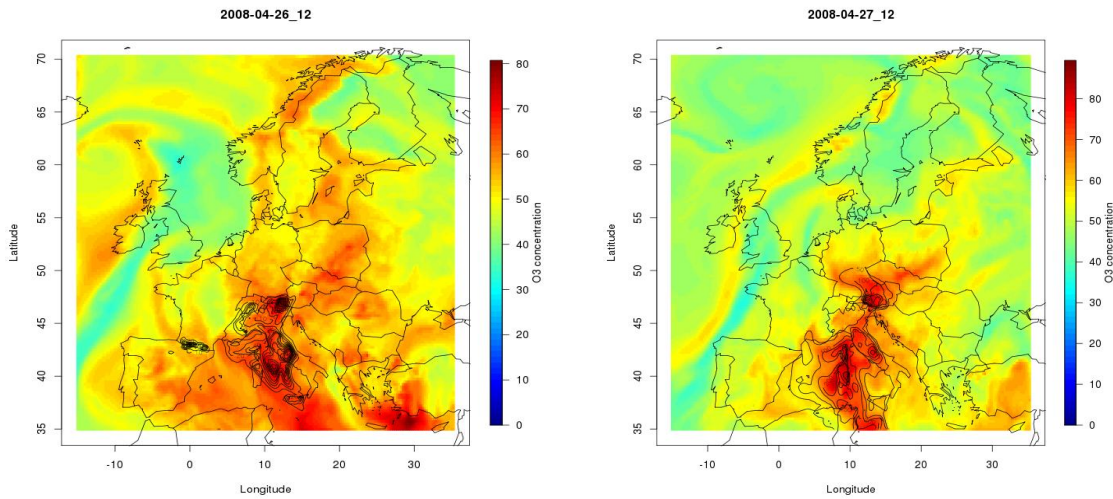
7 **(Lower panel): Hourly average surface ozone (EEA-AirBase) mixing ratios (ppb) at 15:00 h for April 26,**
 8 **2008 (left column) and for April 27, 2008 (right column). Ozone data are from the EEA-AirBase database.**

9

1



2



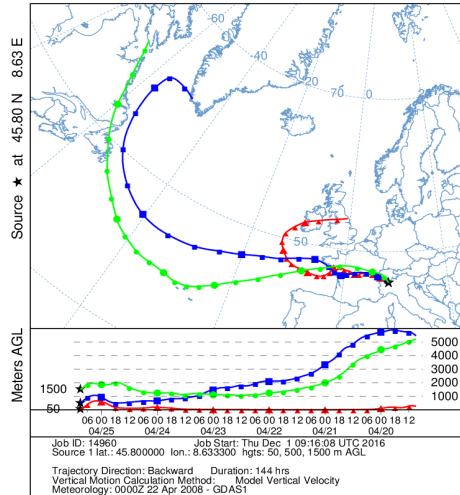
3

4

5

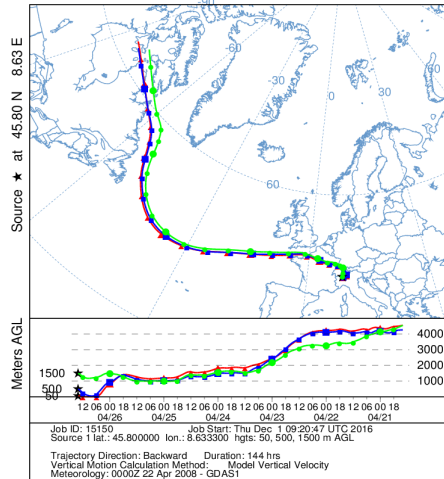
6 **Figure 5** (Upper panel): CHIMERE simulations of the ozone field at 3 km altitude (in ppb) with the iso-
7 contours of the high tropospheric tracer (in black, arbitrary units) for April 26, 2008 (left column) and for
8 April 27, 2008 (right column). (Lower panel): Same as in upper panel but for 1.5 km altitude.

NOAA HYSPLIT MODEL
 Backward trajectories ending at 0900 UTC 25 Apr 08
 GDAS Meteorological Data



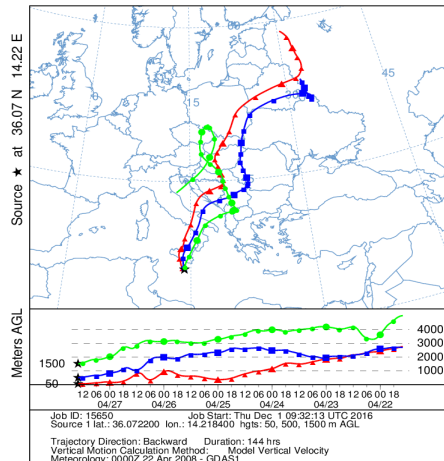
1

NOAA HYSPLIT MODEL
 Backward trajectories ending at 1400 UTC 26 Apr 08
 GDAS Meteorological Data



2

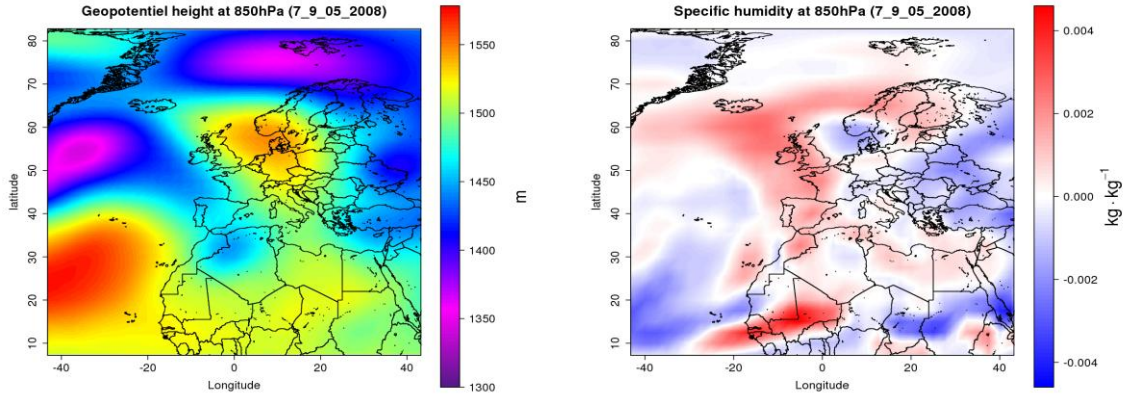
NOAA HYSPLIT MODEL
 Backward trajectories ending at 1400 UTC 27 Apr 08
 GDAS Meteorological Data



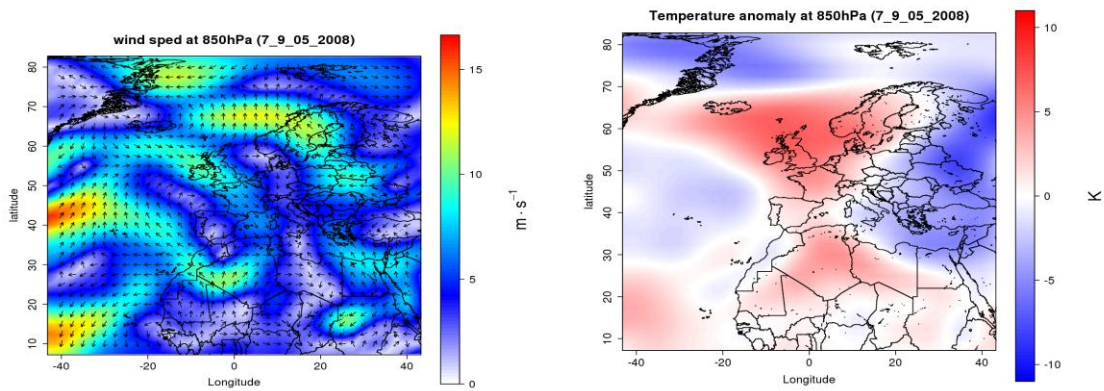
3

4 **Figure 6:** Backward trajectories during the April 25-27 episode ending at EMEP rural ozone stations in
 5 Italy (IT04, upper and middle panels) and Malta (MT01, lower panel).

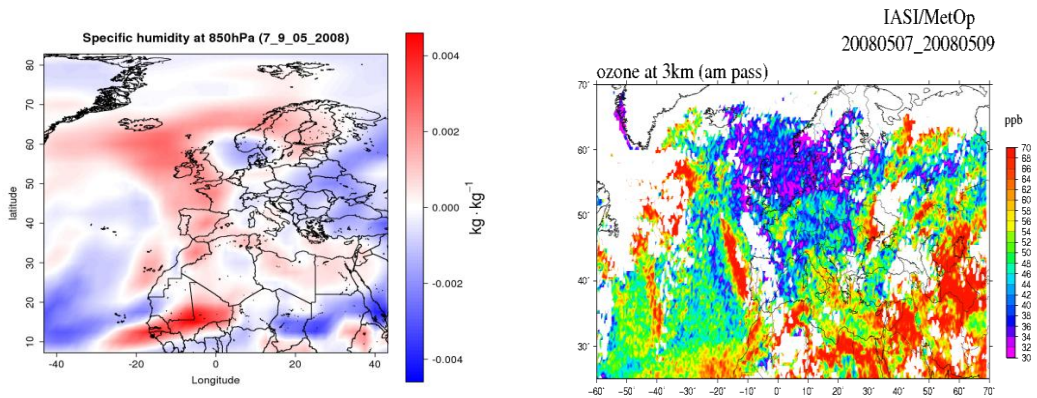
6



1



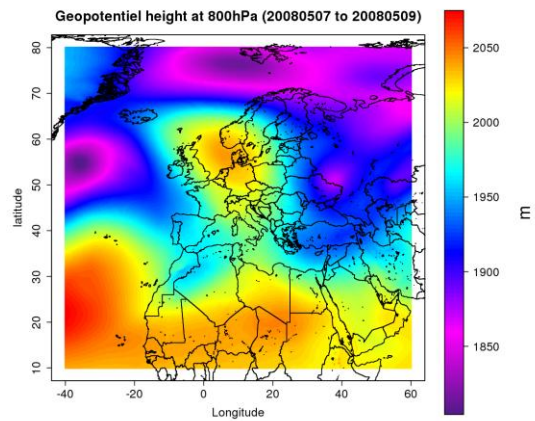
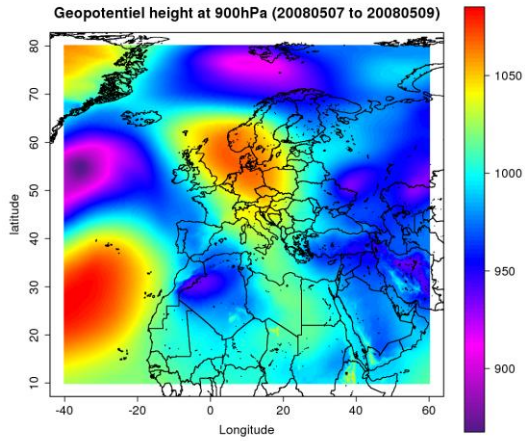
2



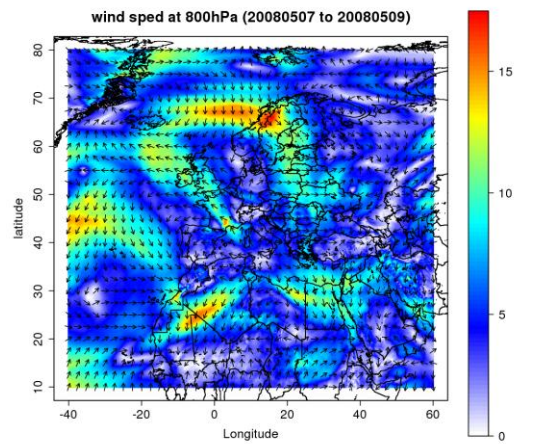
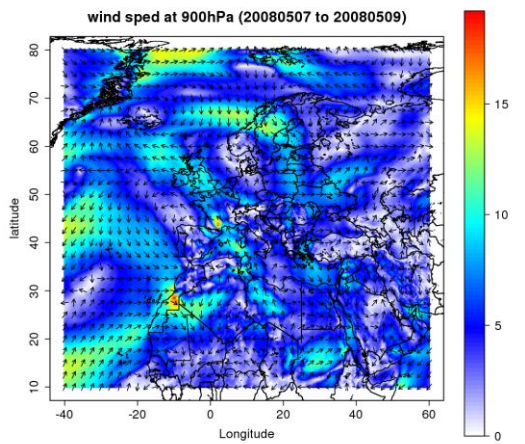
3

4 **Figure 7: Composite NOAA/ESRL weather maps of geopotential height (upper panel-left, in m), specific**
 5 **humidity anomaly (upper panel-right, in Kg/Kg), vector wind (middle panel- left, in m/s) air temperature**
 6 **anomaly (middle panel- right, in K), omega vertical velocity (lower panel-left, in Pa/s) and IASI satellite**
 7 **ozone measurements at 3km (lower panel-right, in ppb) for the high ozone episode of 7-9 May 2008.**

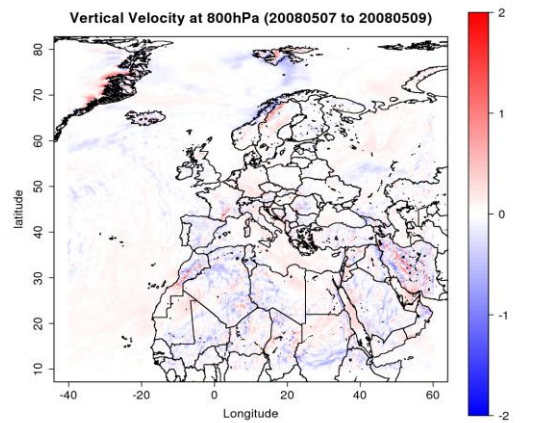
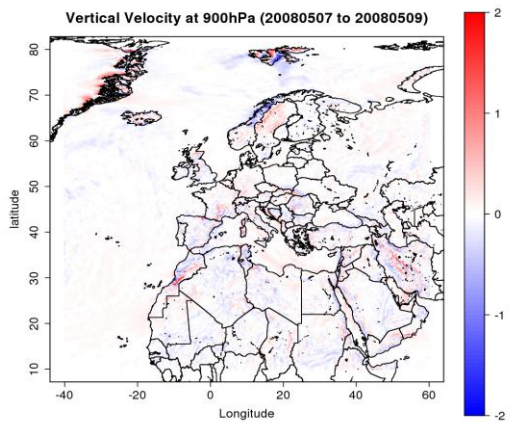
8



1



2

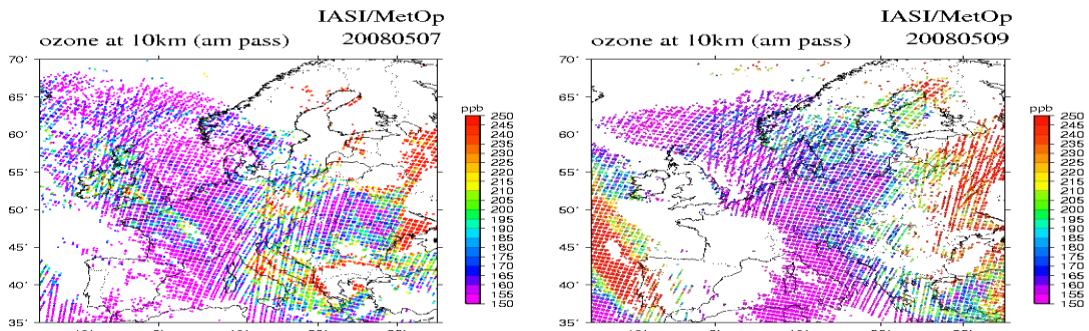


3

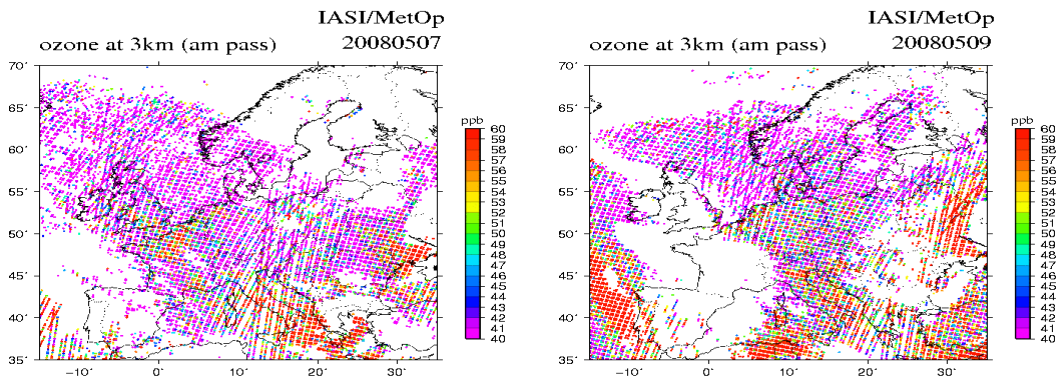
4 **Figure 8: Composite ECMWF meteorological charts of geopotential height (upper panel, in m), wind**
 5 **speed (middle panel, in m/s) and vertical velocity (lower panel, in Pa/s) at 900hPa (left) and 800 hPa**
 6 **(right) for the high ozone episode of 7-9 May 2008.**

7

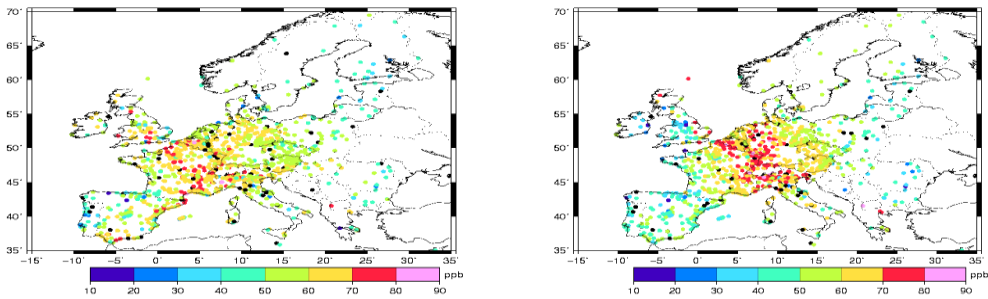
1



2



3



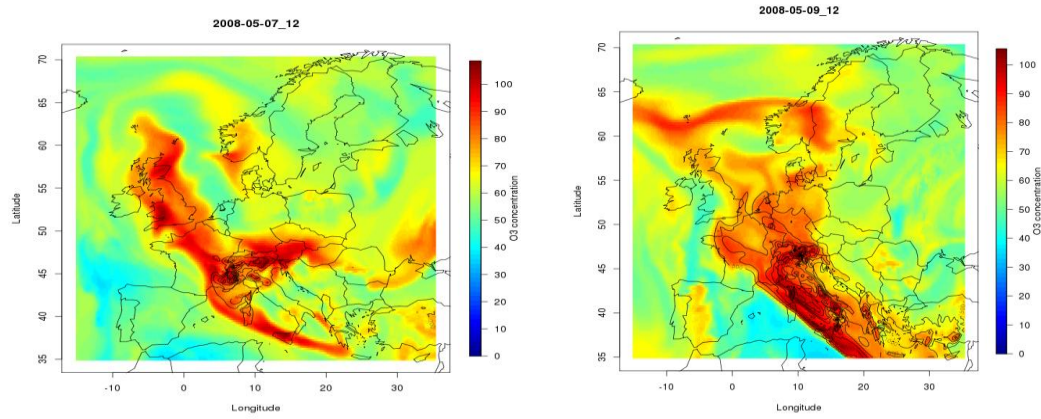
4

5 **Figure 9: Daily IASI satellite ozone measurements at 10 km level (upper panel, in ppb) and at 3km level**
 6 **(middle panel, in ppb) for May 7, 2008 (left column) and May 9, 2008 (right column). Values outside the**
 7 **scale range are set up to the upper and lower color code respectively.**

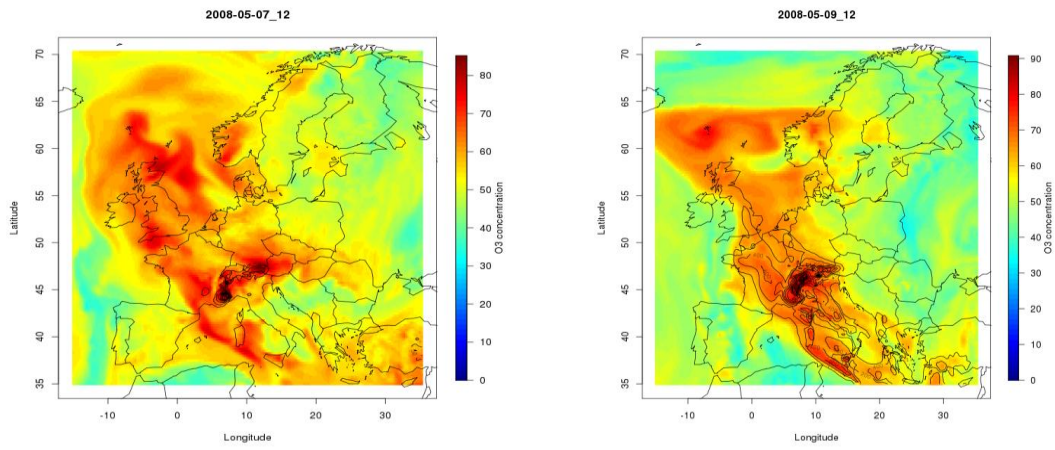
8 **(Lower panel): Hourly average surface ozone (EEA-AirBase) mixing ratios (ppb) at 15:00 h for May 7,**
 9 **2008 (left column) and May 9, 2008 (right column). Ozone data are from the EEA-AirBase database.**

10

1
2



3



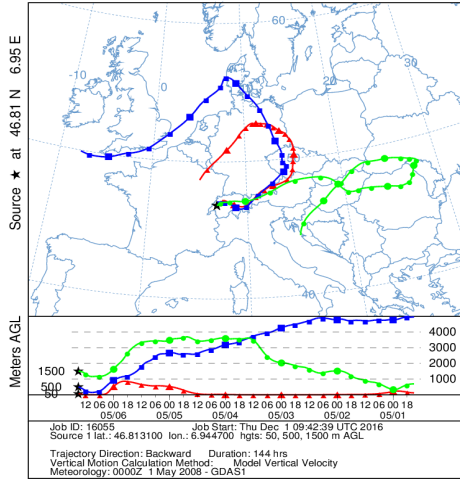
4

5

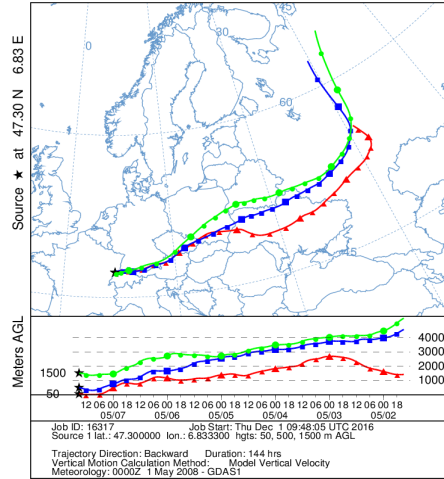
6 **Figure 10** (Upper panel): CHIMERE simulations of the ozone field at 3 km altitude (in ppb) with the iso-
7 contours of the high tropospheric tracer (in black, arbitrary units) for May 7, 2008 (left column), and May
8 9, 2008 (right column). (Lower panel): Same as in upper panel but for 1.5 km altitude.

9

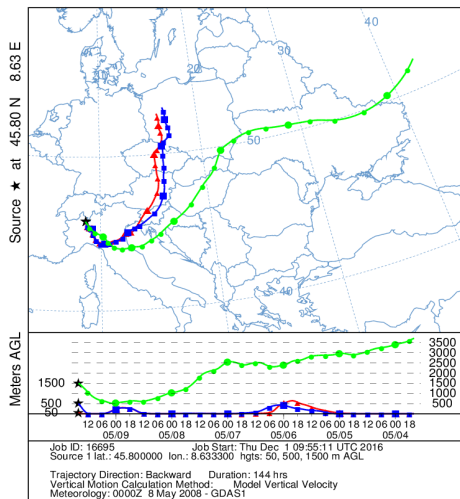
NOAA HYSPLIT MODEL
Backward trajectories ending at 1500 UTC 06 May 08
GDAS Meteorological Data



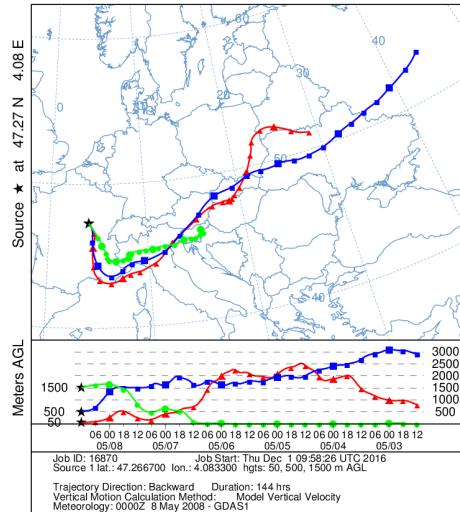
NOAA HYSPLIT MODEL
Backward trajectories ending at 1500 UTC 07 May 08
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 1600 UTC 09 May 08
GDAS Meteorological Data



NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 08 May 08
GDAS Meteorological Data



1

2

3

4

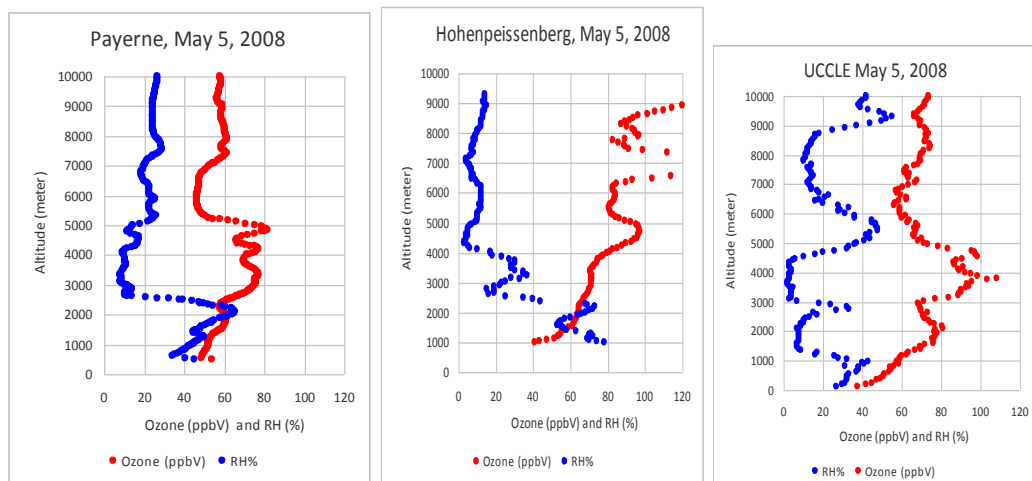
5

6

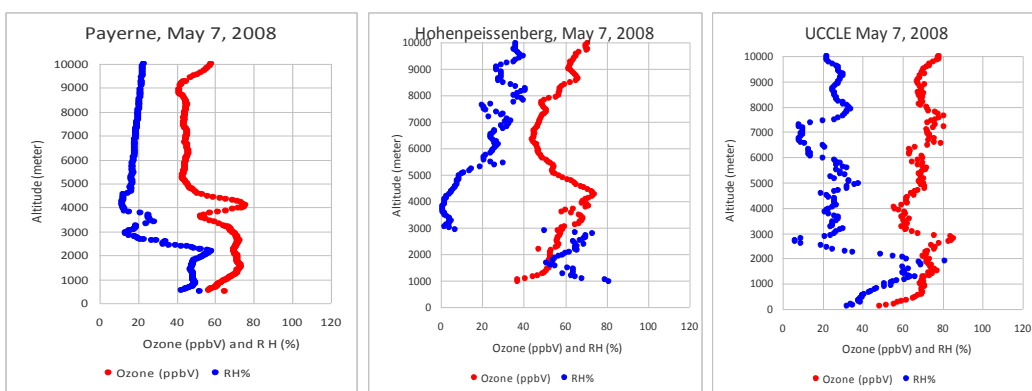
7

Figure 11: Backward trajectories during the 6-9 May 2008 ozone episode ending at EMEP rural ozone stations in Switzerland and Italy (left column) and France (right column).

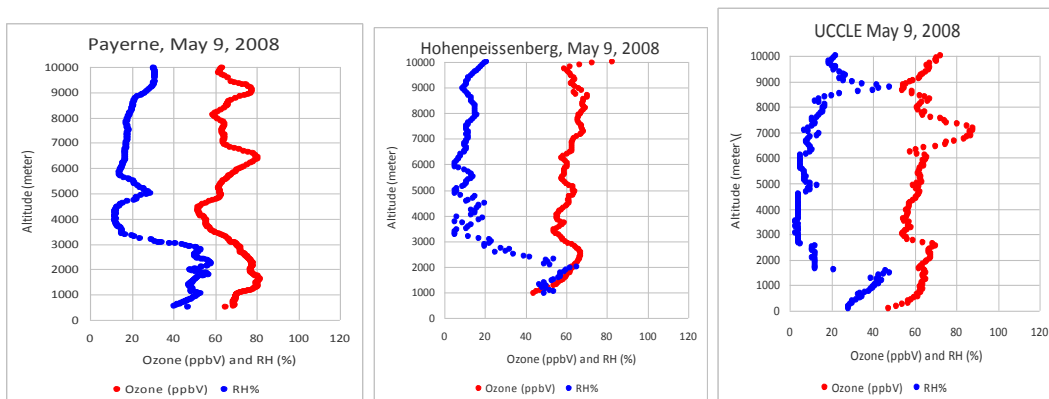
1



2



3



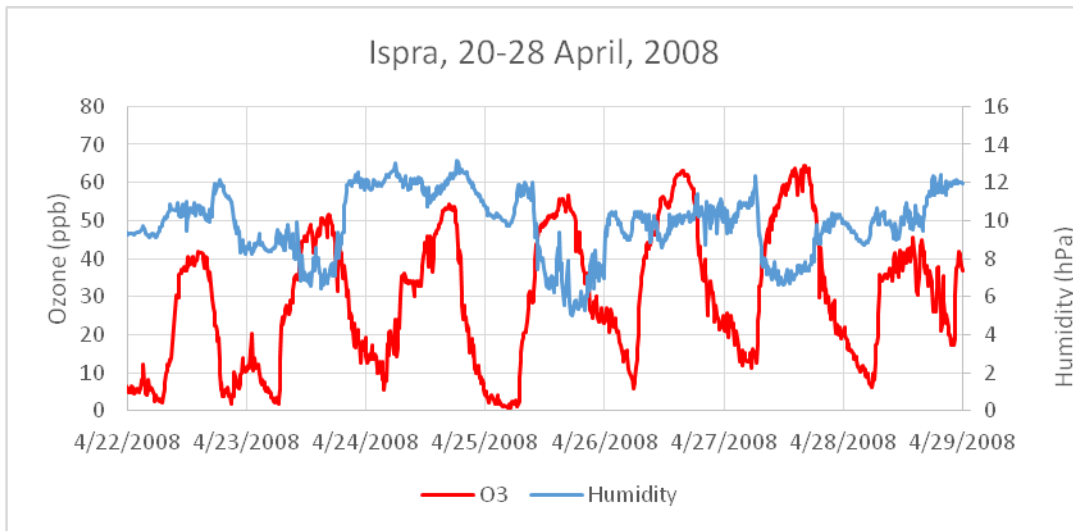
4

5 **Figure 12:** Vertical profiles of ozone (red, ppb) and relative humidity (blue, %) over Payerne, Switzerland
 6 (left column), Hohenpeissenberg, Germany (middle column) and Uccle, Belgium (right column) on May 7,
 7 2008 (upper panel), May 8, 2008 (middle panel) and May 9, 2008 (lower panel).

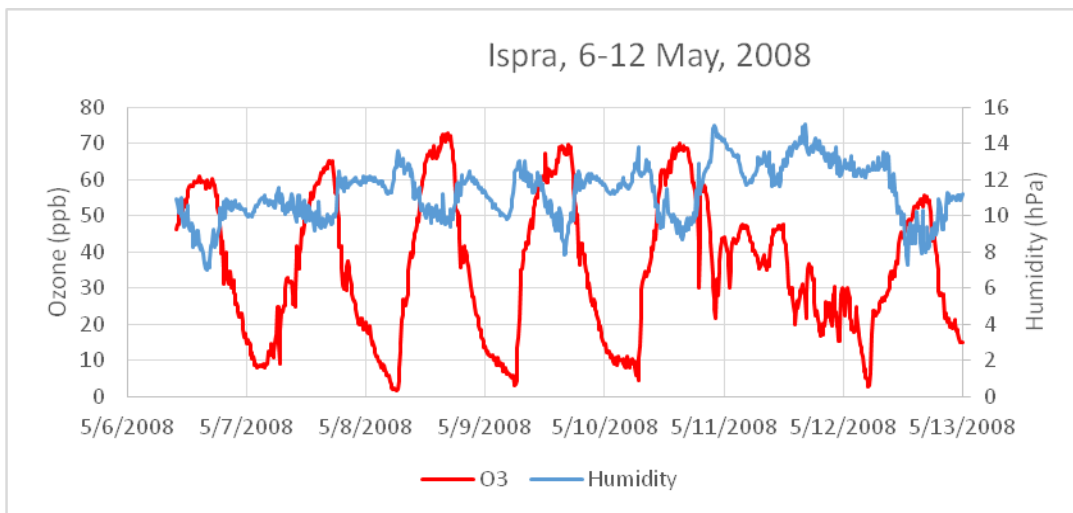
8

9

10



1



2

3

4 **Figure 13:** (Upper panel): Ozone concentration (red, ppb) and absolute humidity (blue, hPa)
 5 measurements at the JRC-Ispra station during the 26-27 April 2008 ozone episode.

6 (Lower panel): Ozone concentration (red, ppb) and absolute humidity (blue, hPa) measurements at the
 7 JRC-Ispra station during the May 7-9, 2008 ozone episode.

8

9

10

11