Anonymous Referee #1 Comments (Authors Response in Italics)

This paper investigates the role of tropospheric subsidence on near-surface ozone concentrations of the JRC-Ispra monitoring station. The study is based on the analysis of several observational products (ground measurements, satellite retrievals), reanalysis data and back trajectories. This is an interesting and solid study, with adequate discussion of the results. However, I think that the structure and number of figures should be revised in order to be more reader-friendly. I recommend publication of the paper after the following comments are addressed.

We would like to thank the reviewer for his positive comments on the manuscript. As seen below we revised the paper according to his suggestions.
The corresponding changes in the revised manuscript (in Final Response) are highlighted in yellow color.

Main comments
a. The manuscript includes too many figures. For example, the third examined episode is associated with 10 figures, while the first episode with only one. My suggestions on this are the following:
1. Merge Figure 1 and Figure 2 into one Figure. Increase the size of the legends in Figure 1 and Figure 2.

Figs 1 and 2 have been merged into one Figure (new Fig. 1) and the size of the legends has been increased.

2. As Figure 6 and Figure 7 are referring to the same episode you can merge them into one Figure. The same stands for Figure 12 and Figure 13.

Figs 6 and 7 have been merged into one Figure (new Fig. 5) as well as Figs 12 and 13(new Fig. 10)

3. As Figures 16a and 16b are of the same temporal extent they can be combined into one figure with a secondary vertical axis. The same stands for Figure 18a and 18b. Then the new fig.16, fig. 17a, fig 17b, and the new fig18 can be merged into one Figure.

Figs 16a, 16b, 17a, 17b, 18a, 18b, have been merged into one Figure (new Fig. 13)

4. Please use a, b, c... labelling for all your Figures.
5. For every Figure use one caption describing there every a, b, c. . . subfigure.

In the new Figures 1 and 13, presenting the JRC-station measurements, the labels a, b, c, d were used and also one caption was used describing the a, b, c, d subfigures.

6. In Figures 3-11, 14 and 15 remove the surrounding white space to improve both the quality and visibility of the figures.

For the indicated Figs the surrounding white space was removed.

b. Regarding the selection of the episodes the authors state that “. . .the 3 most characteristic of them..” will be presented (page 7, line 32). Most characteristic in terms of what? Can you be more specific on this somehow subjective criterion?

The sentence was rephrased as follows:
“More than ten 7Be - ozone weekly episodes were identified in the whole time series and the three most characteristic of them, for what concerns signs of tropospheric subsidence as observed in the meteorological and air pollution measurements (high 7Be and O3 concentrations combined with positive omega and dry air masses), will be presented in the following paragraphs. The selected episodes were: 3-10 May 2011, 23-29 May 2012 and 28 June – 04 July 2011”.

c. I suggest presenting the time period of each of the three episodes at the beginning of Section 3.2. Then every examined episode can be presented as individual Section 3.2.1, 3.2.2 and 3.2.3.

Ok

Comments
1. Apart from tropospheric subsidence influencing near-surface ozone concentrations, there are climatological and case studies of stratospheric intrusions affecting near surface ozone concentrations for the Mediterranean region (Cristofanelli et al., 2006; Gerasopoulos et al., 2006; Akritidis et al., 2010). I believe that the contribution of such events on near-surface ozone for the Mediterranean region should be also included in the Introduction.

The contribution of stratospheric ozone intrusions affecting near surface ozone concentrations for the Mediterranean region has been more stressed in the introduction and the suggested relevant references have been included.
So, the following sentence was added:
Apart from tropospheric subsidence influencing near-surface ozone concentrations,

It has to be mentioned also that there are climatological and case studies of stratospheric intrusions affecting near surface ozone concentrations for the Mediterranean region (Cristofanelli et al., 2006; Gerasopoulos et al., 2006; Akritidis et al., 2010)

2. Page 6, line 18: Please add a reference for the use of the 7Be/210Pb ratio and a small description for the purpose of its usage.

We think that this information is already presented sufficiently in the introduction (Page 3, lines 1-24), where many relevant publications on the use of radionuclide measurements for atmospheric transport are cited. We added also a recent relevant paper, which describes very extensively the use of the 7Be and 210Pb radionuclides in atmospheric transport studiesin its introduction (Brattich et al., 2017). The referenced WMO-GAW report (2004) as well as the cited reference Koch et al., 1996 make also an interesting review on this subject.
In addition, the following phrase was added in the 1st line of the respective paragraph in the introduction:
"and in particular terrigenous 210Pb and cosmogenic 7Be, which are natural radionuclides that are helpful in understanding the roles of transport and/or scavenging in controlling the behavior of radiatively active trace gases and aerosol"

Technical comments
1. Page 2, line 7: Remove dot after "are observed."
Ok

2. Page 3, line 6: Replace "if 3.8 days" with "of 3.8 days".
Ok

3. Page 3, lines 12-13. Please correct the order of references. Also, check for other similar instances throughout the manuscript and correct accordingly.
Ok

4. Page 3, line 32: Replace "during summertime ozone episodes over the eastern Mediterranean and linked" with "during the summertime ozone episodes over the eastern Mediterranean and are linked".
Ok

5. Page 4, line 21 to Page 5 line 4. I suggest including bullets for the description of the measurements.
Ok

6. Page 4, line 22: Replace "Jensen et al., 2017" with "Jensen et al. (2017)". Also, check for other similar instances throughout the manuscript and correct accordingly.

Ok

7. Page 4, line 27: Delete the extra dot.
Ok

8. Page 5, line 6: Replace “charts for” with “charts of”.
Ok

9. Page 5, line 7: Replace “for the atmospheric “with “at the atmospheric”.
Ok

10. Page 6, line 17: Replace “and of ozone vs” with “and that of ozone vs”.
Ok

11. Page 7, lines 31-32: Replace 10 and 3 with “Ten” and “three”.
Ok

12. Page 11, lines 13-17. This is a rather long sentence. Please rephrase.

The sentence was modified as follows:
“During high 7Be and high ozone episodes, the highest evening ozone values exceeding the standards usually occur within the following 2-3 days after the maximum of regional tropospheric subsidence, as observed also in the analysis of several episodes not presented in this paper. The increase in ozone concentrations usually occurs under the influence of favourable meteorological conditions for photochemical ozone production in the boundary layer, which is added-up on the increased regional background due to tropospheric subsidence and thus occasionally leading to exceedances in ozone air quality standards”.

13. Page 32, Figure 10: Please rephrase the figure caption to be clearer.

The figure caption was rephrased.

References

Akritidis, D., Zanis, P., Pytharoulis, I., Mavrakis, A., and Karacostas, T.: A deep stratospheric intrusion event down to the earth’s surface of the megacity of Athens, Meteorol. Atmos. Phys., 109, 9–18, 2010
Cristofanelli, P., Bonasoni, P., Tositti, L., Bonafe, U., Calzolari, F., Evangelisti, F., Sandrini, S., and Stohl, A.: A 6-year analysis of stratospheric intrusions and their influence on ozone at Mt. Cimone (2165m above sea level), J. Geophys. Res.-Atmos., 111, D03306, https://doi.org/10.1029/2005JD006553, 2006.
Gerasopoulos, E., Zanis, P., Papastefanou, C., Zerefos, C. S., Ioannidou, A., and

Wernli, H.: A complex case study of down to the surface intrusions of persistent stratospheric air over the Eastern Mediterranean, Atmos. Environ., 40, 4113–4125, 2006.

Anonymous Referee #2 Comments (Authors Response in Italics)

The article presents specific case studies to interpret the role of subsidence on high elevation surface ozone concentrations, with the synergy of different in situ and satellite observational data. Overall, the paper merits publication after a number of comments are taken into account:

We would like to thank the reviewer for his positive comments on the manuscript. As seen below we revised the paper according to his suggestions.
The corresponding changes in the revised manuscript (in Final Response) are highlighted in green color.

In the abstract, a 10 year measurement period is mentioned, which creates a clear expectation that the results of this study are put in a long term context. This is not happening and it is a clear gap of the study. Unless the criteria used to select the cases are analysed in a previous paper (in that case direct reference to result from that paper(s) should be included), the authors should provide some statistics on the extent at which their findings for 2011 are also typical for the rest of the years as well as some means of quantification (e.g. frequency, values during events versus average values).

We would like to emphasize that the study is focused on atmospheric mechanisms, based on some selected case-studies and it is not a statistical one. In combination with a similar comment of reviewer 1 the relevant phrase (page 8, lines 3-7) was modified as follows "More than ten 7Be - ozone weekly episodes were identified in the whole time series and the three most characteristic of them, for what concerns signs of tropospheric subsidence as observed in the meteorological and air pollution measurements (high 7Be and O3 concentrations combined with positive omega and dry air masses) will be presented and examined in the following paragraphs. The selected episodes were: 3-10 May 2011, 23-29 May 2012 and 28 June – 04 July 2011. The episodes discussed here are not Foehn events". In addition, the reader might have an idea of the frequency of occurrence of these episodes by having a look at Fig. 1 as well as in the Supplement Figs S1-S4 where the weekly averages of 7Be and O3 concentrations for 5 years (2006, 2007, 2008, 2011, 2012) plotted. As observed, during the April – September period about 2-3 major 7Be - O3 episodes are spotted. As mentioned in the manuscript, these episodes could be better detected with shorter than weekly measurements of 7Be as the usual duration of subsidence episodes is about 2-3 days (see new Fig. 13), but such measurements were not available.

The amounts of plots used is huge! The authors should definitely make a serious attempt either to merge few of them, or move to suppl. material, or exclude if really not needed. Those changes might need to be followed by changes in the text and the overall structure, which I leave upon the authors, yet few suggestions will follow immediately after in my review.

A significant merging and reduction of plots associated with the corresponding text changes have been undertaken in combination also with similar comments of reviewer 1, leading to a total number of 13 Figures in the revised text.

Finally, there are parts of the introduction or the results where references do not seem to be up to date, either in terms of time or space, the latter meaning references relevant to the area of interest. I have included few examples which I consider only indicative, but a more through review of the current state might be needed, and the selection remains at the discrete consideration of the authors.

Following the reviewer's comment an update of references has been made in the introduction and the results section, as it is described in the following paragraphs.

Specific comments:
Pg 1, Ln 30 – "It has been reported that tropospheric . . . the last couple of centuries (Volz and Kley, 1988; Forster et al., 2007)." I would suggest that this introductory statement should be supported with more recent references.

Following the reviewer's suggestion two more recent references, which are review papers on tropospheric ozone were mentioned for supporting the introductory statement (Monks et al., 2015; Gaudel et al., 2018).

Pg2, Ln 6 – "which might also be associated to deep tropospheric subsidence especially over the Mediterranean . . . Kalabokas et al., 2013; Cooper et al., 2014; Safieddine et al., 2014; Kalabokas et al., 2015) . . . especially for deep stratospheric intrusions the following references are very characteristic for the area and should be included in the already too long list of references, or later (Pg 2, lines 20-25). A deep stratospheric intrusion event down to the Earth's surface of the megacity of Athens April 2012 Meteorology and Atmospheric Physics 109(1):9-18, DOI: 10.1007/s00703-010-0096-6 by Akritidis et al. Gerasopoulos E, Zanis P, Papastefanou C, Zerefos CS, Ioannidou A,Wernli H (2006) A complex case study of down to the surface intrusions of persistent stratospheric air over the EasternMediterranean. Atmos Environ 40:4113–4125. Kentarchos AS, Davies TD, Zerefos C (1998) A low latitude stratospheric intrusion associated with a cut-off low. Geophys. Res. Lett. 25:67–70

Following the reviewer's suggestion, the mentionned references on deep tropospheric subsidence over the Mediterranean were added into the text.

Pg 2, Ln 31 – It seems that two studies conducted at Finokalia remote station in the eastern Mediterranean, dealing with the dynamics and photochemistry of ozone are missing from the introduction, especially when discussing the eastern Mediterranean controlling mechanisms of surface ozone. On the contrary, there are many selfcitations from the first author that need to be enriched with studies from other groups in the area. Kouvarakis, G., K. Tsigaridis, M. Kanakidou, and N. Mihalopoulos (2000), Temporal variations of surface regional background ozone over Crete Island in the southeast Mediterranean, J. Geophys. Res., 105(D4), 4399 – 4407. Photochemical ozone production in the Eastern Mediterranean, June 2006, Atmospheric Environment 40(17):3057-3069, DOI: 10.1016/j.atmosenv.2005.12.061 by Gerasopouolos et al. Gerasopoulos, E., G. Kouvarakis, M. Vrekoussis, M. Kanakidou, and N. Mihalopoulos (2005), Ozone variability in the marine boundary layer of the eastern Mediterranean based on 7-year observations, J. Geophys. Res., 110, D15309, doi:10.1029/2005JD005991.

The suggested studies on surface ozone conducted at the Finokalia station were added into the manuscript.

Pg 3, Ln 15-17: Be7 reference for ambient levels are quite old, some inquiry on new articles reporting on the levels should be done, especially in the area of interest. The same in lines 21-23.

Following the reviewer's suggestion ten more recent references on 7Be ambient levels were added, especially concerning the area of interest of the study (European Continent – Western Mediterranean): Bourcier et al., 2011; Brattich et al 2017; Duenas et al, 2011; García et al, 2012; Hernández-Ceballos et al, 2016; Ioannidou et al, 2014; Jiwen et al, 2013; Leppanen et al, 2010; Lozano et al, 2012; Pham et al, 2011; Steinmann et al, 2013.

The information in section 2.2 should be better included in a table.

A table has been added to include the information in section 2.2 (Instrumentations and measurements at JRC-Ispra site).

Figure 1a could be combined with 1b. The same stands for 2a, 2b.

The suggested combination of Figs has been done following also a similar comment from reviewer 1.

Overall, the added value of this paper results is not clear and should be better highlighted, mostly in the conclusions. It is obvious that it is an extension of previous works and for that reason it needs to be clear where does this study starts from and where it ends up (added value) at the same time being a self standing scientific publication.

In the previous study (Kalabokas et al., 2017) some troposheric mechanisms related with regional ozone episodes especially linked with large-scale subsidence were examined mainly based on surface ozone, IASI vertical columns and meteorological analysis. In this manuscript a more detailed analysis of the suggested mechanisms was performed, based on the measurements of a very large variety of meteorological and air pollution parameters collected at the JRC-Ispra station, which is considered as one of the most well-equipped measuring sites in Europe. This measurement set includes tracers of both subsidence (^{7}Be, RH), boundary layer origin (^{222}Rn, ^{210}Pb, NOx) and photochemical activity (partly PM), and this allows at least qualitatively distinguish origin of different air masses and trace back ozone origin. Relevant phrases have been inserted at the last paragraph of the introduction as well as at the beginning of the conclusions section.

A study of the influence of tropospheric subsidence on spring and summer surface ozone concentrations at the JRC-Ispra station in northern Italy

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Abstract. The influence of tropospheric ozone to the surface ozone concentrations is investigated at the monitoring station of JRC-Ispra, based on 10 years of measurements (2006-2015) of surface ozone data. In-situ hourly measurements of ozone and other air-pollutants, meteorological parameters and weekly averaged ^{7}Be (as indicator of upper-tropospheric/stratospheric influence) and ^{210}Pb measurements (as indicator of boundary layer influence), have been used for the analysis. In addition, IASI+GOME2 and IASI ozone satellite data have also been used. It is observed that frequently ^{7}Be and ozone weekly peaks coincide, which might be explained by the impact of deep atmospheric subsidence on surface ozone, particularly during late spring and early summer. Based on this observation, a detailed analysis of selected ^{7}Be and ozone episodes occurring during that period of the year has been performed in order to further elucidate the mechanisms of tropospheric influence to the surface pollutant concentrations. For the analysis composite NOAA/ESRL reanalysis synoptic meteorological charts in the troposphere have been used as well as IASI satellite ozone measurements and NOAA-HYSPLIT back trajectories. The JRC-station hourly measurements during subsidence episodes show very low values of local pollution parameters (e.g. NO_x, ^{222}Rn, nephelometer data, PM_{10}), close to zero. On the contrary, during these periods ozone levels reach usually values around 45-60 ppb during the afternoon hours but also show significantly higher values than the average during the night and morning hours, which is a sign of direct tropospheric influence to the surface ozone concentrations.

1 Introduction

It has been reported that tropospheric ozone levels as well as surface ozone concentrations have increased significantly during the last couple of centuries (Volz and Kley, 1988; Forster et al., 2007; Monks et al., 2015; Gaudel et al., 2018). Ozone is an important greenhouse gas and might cause adverse effects to human health and also have negative impacts on vegetation and materials (Ito et al., 2005; Van Dingenen et al., 2009; Hollaway et al., 2012). Tropospheric ozone is mainly produced in the troposphere through photochemical reactions of precursor pollutants but it does also originate from stratospheric intrusions (Volz and Kley, 1988; Staehelin et al., 1994). On the average, the tropospheric ozone originating from the stratosphere is about 10 % on global scale (Monks et al., 2015). The surface ozone concentrations depend on photochemistry and transport within the boundary layer as well as tropospheric entrainment, which might also be associated to deep tropospheric subsidence especially over the Mediterranean during summer where, in general, high ozone levels are observed (Kalabokas et al., 2000; Kouvarakis et al., 2000; Lelieveld et al., 2002; Zerefos et al., 2002; Gerasopoulos et al., 2005; Kalabokas et al., 2007; Kalabokas et al., 2008; Lelieveld et al., 2009; Velchev et al., 2011, Richards et al., 2013; Kalabokas et al., 2013; Cooper et al., 2014; Safieddine et al., 2014; Kalabokas et al., 2015).

The influence of intercontinental ozone transport on surface ozone concentrations is considered as a critical issue regarding ozone pollution control (UNESCE, 2010) and this issue has been extensively studied especially in the USA where many studies have shown that tropospheric transport and entrainment of ozone from the free troposphere to the boundary layer has an important impact on surface ozone-mixing ratios (Cooper et al., 2011; Cooper et al., 2012; Parrish et al., 2013; Parrish et al., 2014; Langford et al., 2015). Also, anticyclonic synoptic conditions are normally linked to tropospheric subsidence, which is considered to be an important source of high ozone concentrations over the eastern Mediterranean. During the summer period, the Mediterranean area is under the influence of the descending branch of the Hadley circulation (Lelieveld, 2009) in combination with the impact of the Indian monsoon, inducing a Rossby wave that through the interaction with the mid-latitude westerlies produces adiabatic descent in the area (Rodwell and Hoskins, 1996, 2001; Tyrlis et al., 2013). In the eastern Mediterranean strong deep subsidence throughout the troposphere to the boundary layer, which seems to be a quite frequent phenomenon, has been documented based on the analysis of MOZAIC vertical ozone profiles as well as surface ozone and satellite measurements (Kalabokas et al., 2007, 2008, 2013, 2015; Eremenko et al., 2008; Foret et al., 2009; Liu et al., 2009; Doche et al., 2014; Gaudel et al., 2018). In addition, large-scale atmospheric modeling simulations (Li et al., 2001; Richards et al., 2013; Zanis et al., 2014; Safieddine et al., 2014; Tyrlis et al., 2014; Akritidis et al., 2016; Cristofanelli et al., 2018) show the importance of the vertical ozone transport over the Mediterranean basin, especially in its eastern side. Also, sea-breeze circulation appears to have a particularly strong influence on ozone formation in the western Mediterranean, because it favors accumulation of ozone in recirculated polluted air masses (Millan et al. 1997, 2000; Castell et al. 2008). The atmospheric processes controlling ozone levels over the western and central Mediterranean need further studying, especially for what concerns the springtime months. It has been reported that surface background ozone levels in the western and central Mediterranean show a maximum in spring (April–May) while in the eastern Mediterranean stations a later ozone maximum appears in July–August (Kalabokas et al., 2008; Zanis et al., 2014). Also, it has been recently reported that high spring-time ozone concentrations are detected over a large geographical area for several days under specific synoptic conditions, which could be explained by the impacts of tropospheric subsidence (Kalabokas et al., 2017). Apart from tropospheric subsidence influencing near-surface ozone concentrations, it has to be mentioned also that there are climatological and case studies of stratospheric intrusions affecting near surface ozone concentrations for the Mediterranean region (Kentarchos et al., 1998; Cristofanelli et al., 2006; Gerasopoulos et al., 2006; Akritidis et al., 2012).

Atmospheric radionuclides are useful for studies of tropospheric subsidence and transport and in particular terrigenous ^{210}Pb and cosmogenic ^{7}Be, which are natural radionuclides that are helpful in understanding the roles of transport and/or scavenging in controlling the behavior of radiatively active trace gases and aerosol (WMO-GAW, 2004; Feichter et al., 1991; Balkanski et al., 1993; Koch et al., 1996; Graustein and Turekian, 1996; Arimoto et al., 1999; Gerasopoulos et al., 2001; Zanis et al., 2003; Liu et al., 2004; Leppanen et al., 2010; Pham et al., 2011; Garcia et al., 2012; Jiwen et al., 2013; Cuevas et al., 2013; Ioannidou et al., 2014; Brattich et al., 2017). Due to their different origins, the investigation of ^{210}Pb and ^{7}Be activities simultaneously can be useful for studies of atmospheric transport of pollutants, especially in particle phase (Koch et al., 1996). ^{210}Pb has a half-life of about 22 years. It originates from the decomposition of ^{222}Rn, which has a half-life of 3.8 days and it is a decomposition product of ^{226}Ra originating from the ground (Baskaran, 2011). ^{7}Be has a cosmogenic origin and it is formed mostly by the decomposition of the atoms of carbon, nitrogen and oxygen present in the atmosphere by incident gamma radiation. It has a half-life of 53.3 days (Masarik and Beer, 1999). Its production rate increases with altitude and saturates at about 15 km height (Usokin and Kovaltsov, 2008). Most of ^{7}Be is produced in the stratosphere and about the one third in the troposphere, especially in its upper part (O'Brien, 1979). After their formation, ^{7}Be atoms are mostly attached to atmospheric particles and so their atmospheric concentrations are greatly influenced by transport and deposition processes of particles (Jaenicke, 1988; Feely et al., 1989; Papastefanou and Ioannidou, 1995; Koch et al., 1996; Bourcier et al., 2011; Duenas et al., 2011; Lozano et al., 2012; Steinmann et al., 2013; Hernández-Ceballos et al., 2016).

On the average, the ^{7}Be concentrations in the upper troposphere are about the 25% of the lower stratospheric concentrations (about160 mBq m^{-3}) at northern mid-latitudes while ^{7}Be concentrations close to the ground are generally below 5 mBq m^{-3}

(Reiter et al., 1983; Dutkiewicz and Husain, 1985; Brost et al., 1991; Gaggeler, 1995). So, air masses originating from the upper troposphere or stratosphere contain usually high ^{7}Be concentrations and the intrusions of stratospheric air mass into the troposphere are the main processes transferring ^{7}Be to the earth's surface through dry or wet deposition. Therefore, ^{7}Be serves as a tracer for lower stratospheric and upper tropospheric air masses arriving to the ground (Lee et al., 2007; Papastefanou et al., 2012). Due to the exchange and removal processes in the atmosphere the ^{7}Be concentration in air at ground level varies strongly with the season while ^{7}Be production rates in the atmosphere remain relatively constant (Durana et al., 1996; Masarik and Beer, 1999). In general, ^{7}Be surface concentrations show a maximum in late summer (Reiter et al., 1983; Feely et al., 1989; Bourcier et al., 2011; Hernández-Ceballos et al., 2016).

The investigation of the atmospheric processes controlling the frequently observed springtime ozone maximum over parts of the European continent including the western Mediterranean is an interesting research issue as photochemical ozone production and tropospheric transport, including stratospheric influence, might be involved (Beekmann et al., 1994; Monks, 2000). It has been previously reported that high surface ozone concentrations, lasting several days, have been observed over large geographical areas at the same time (Kalabokas et al., 2017). It has been also shown that the observed regional springtime ozone episodes are usually associated to specific synoptic meteorological patterns, which have great similarities with those observed during the summertime ozone episodes over the eastern Mediterranean and are linked to large-scale subsidence (Kalabokas et al., 2013, 2015; Doche et al., 2014).

In the previous study (Kalabokas et al., 2017) some troposheric mechanisms related with regional ozone episodes especially linked with large-scale subsidence were examined. In this manuscript a more detailed analysis of the suggested mechanisms was performed, based on the measurements of ^{7}Be , ^{210}Pb as well as a large variety of meteorological and air pollution parameters collected at the JRC-Ispra station, which is considered to be one of the most well-equipped measuring sites in Europe. The present study focuses on the influence of tropospheric subsidence on surface ozone concentrations, especially during spring and summer months over the western Mediterranean area. It is based on the analysis of 10 years of ozone and other air pollution measurement data (2006-2015) as well as measurements of natural radionuclide tracers at surface level, which can be used as tracers of transport and photochemical and removal processes. These data are collected at the JRC-Ispra site, located in the pre-alpine area in northern Italy but also located relatively close to the western Mediterranean. In addition, we used meteorological maps, back trajectories and IASI satellite ozone data for a better understanding of the relative importance of the contributions of the different sources of ozone, especially the role of the vertical transport in the troposphere. To our knowledge, a similar analysis of such multi-parameter long term measurements has not yet been performed so far for this area.

2 Experimental

2.1 Site description

The JRC-Ispra station (45.807°N, 8.631°E, 223m a.s.l) is located in a valley in the pre-alpine area of northern Italy. A general meteorological characteristic of the area is that low winds usually prevail with occasional northerly Foehn events (Mira-Salama et al., 2008). More details about the site can be found in Putaud et al., 2017.

2.2 Instrumentations and measurements at JRC-Ispra site

The information on the instrumentation used for this investigation is presented in Table 1.
Regarding the meteorological measurements, a WXT510 weather transmitter from "Vaisala" recorded simultaneously 6 meteorological parameters, temperature, pressure, relative humidity, precipitation, wind speed and wind direction, from the top of an about 10 m high mast. Humidity (in ppmV units) was calculated from relative humidity (RH), temperature and atmospheric pressure, which was useful for the data-analysis as an air mass reaching the surface by transport from higher altitudes will typically be relatively dry and entrainment of air from the free troposphere is thus normally associated with a drop in humidity mixing ratio as well as specific humidity.

2.3 Meteorological maps and back trajectories

Composite NOAA/ESRL reanalysis meteorological charts of various meteorological parameters for selected ozone episodes and at the atmospheric pressure levels of 850, 700 and 500 hPa have been produced with a horizontal resolution of 2.5º × 2.5º (Kalnay et al., 1996). Due to space limitations mostly the 700 hPa charts, representative for the free troposphere, are presented. In addition, 6-day NOAA /HYSPLIT back trajectories for selected high ozone days for air masses arriving at the JRC-Ispra site at various end points have been plotted, using the GDAS meteorological data (Draxler and Rolph, 2015).

2.4 IASI and IASI+GOME2 satellite ozone measurements

Progresses in satellite observations of tropospheric ozone have been made during the last decade (e.g., Fishman et al., 2003; Liu et al., 2005; Coheur et al., 2005; Worden et al., 2007; Eremenko et al., 2008, Cuesta et al., 2013). These progresses combined with their spatial coverage and horizontal resolution make possible to use such observations to complement in situ observations and to support the analysis of ground measurements as well as modeling simulations. In this study, we use ozone satellite observations derived from the IASI infrared instruments and from the multispectral synergism of IASI and GOME-2 in the ultraviolet, for enhancing sensitivity to ozone closer to the surface. The first IASI instrument (Clerbaux et al., 2009) has been launched on board the MetOp-A platform on 19 October 2006. It is a Fourier transform spectrometer operating at nadir in the thermal infrared between 645 and 2760 cm^{-1} with an apodized spectral resolution of 0.5 cm^{-1}. IASI monitor atmospheric composition twice a day at any (cloud-free) location at high resolution with its swath width of 2200 km and its field of view composed of a 2×2 pixels matrix with a diameter at nadir of 12 km each (e.g., Boynard et al., 2009; George et al., 2009; Clarisse et al., 2011; Coman et al., 2012). As in Doche et al. (2014), IASI ozone concentrations retrieved at 3 km and 10 km height are used for our analysis, as representative of the lower and upper troposphere, respectively. The maximum of sensitivity of IASI retrievals ranges between 3 and 5 km in the lower troposphere and 9 and 12 km in the upper-troposphere. Several studies show that the ozone concentrations retrieved from IASI in the lower- and the upper-troposphere are mainly uncorrelated (Dufour et al., 2010, 2012, 2015). We use the vertical profiles retrieved from IASI to calculate longitudinal transect for different latitudes. Vertical profiles within 1° in latitude and 0.5° in longitude are averaged in the transect calculation.

The satellite multispectral approach used here is called IASI+GOME2 (Cuesta et al., 2013). It is based on the joint and simultaneous use of both GOME-2 and IASI measurements for deriving a unique ozone profile for each pair of spectra. It is designed for observing lowermost tropospheric ozone located below 3 km of altitude (with typically a peak of maximum sensitivity down to 2 km of altitude), which is not directly observed with single-band retrievals. As IASI, GOME-2 is only onboard the MetOp satellite series and offer global coverage every day (for MetOp-A around 09:30 local time) with a swath width similar to IASI and a ground resolution moderately coarser than IASI (pixels of 80 km × 40 km for GOME-2). As described in detail by Cuesta et al., (2013), co-located IR and UV spectra are jointly fitted to retrieve a single vertical profile of ozone for each pixel at the IASI horizontal resolution. The UV measurements from the closest GOME-2 pixel (without averaging) are used for each IASI pixel. As for IASI only retrievals, a priori ozone profiles representative of tropical, mid-latitude and polar conditions are calculated by averaging the climatological ozone profiles from McPeters et al., (2007) over the 20-30°N, 30-60°N and 60-90°N latitude bands. The selection of the a priori profiles used during the retrieval is based on the tropopause heights (determined by the temperature vertical profile for each IASI pixel) above 14 km, between 14 and 9 km and below 9 km, respectively. IASI+GOME2 retrievals are routinely produced at the global scale by the French data centre AERIS and they are publicly available (https://iasi.aeris-data.fr/O3_IAGO2/).

3 Results and discussion

3.1 Seasonal variation of O_3 concentrations, ^{7}Be concentrations and the $^{7}Be/^{210}Pb$ ratio.

For the investigation of entrainment episodes and based on information presented in the introduction, plots of weekly averages of ozone vs. ^{7}Be concentrations and that of ozone vs. $^{7}Be/^{210}Pb$ ratios were made for all years during the examined period (2006-2015) and the year 2011 is shown as an example (Figs. 1a,b). As observed, ^{7}Be and O_3 peaks are in several cases coinciding, which might be explained by an impact of deep atmospheric subsidence (air masses moving down from the stratosphere/upper troposphere) on surface ozone. Particularly during springtime, the high ozone levels during such events may be influenced by ozone rich air being transported down to the boundary layer from the stratosphere/upper troposphere.

As seen in Figures 1a-b, in May 2011 and in a period around end of June 2011 and beginning of July 2011 there were episodes of downward transport of ozone down to the surface from the above tropospheric layers. The relatively higher $^{7}Be/^{210}Pb$ ratios at the end of the spring to the beginning of the summer seasons (mid-April to mid-July) indicate that stratospheric or upper tropospheric influence should be most important during this period.

In addition, in Figures 1c-d the ^{7}Be activity and the $^{7}Be/^{210}Pb$ ratio are presented together with specific humidity. As observed, very often the local $^{7}Be/^{210}Pb$ maxima coincide with local minima of specific humidity, which supports the assumption, that this isotope ratio is an indicator of the relative importance of entrainment of subsiding dry air originating from the upper atmospheric into the boundary layer and the ground surface. The ^{7}Be activity does not show a similar correlation with specific humidity. If we look at the yearly variation of the isotope ratio, it has a maximum in the early summer while specific humidity has a maximum later in the summer, which may be explained by the fact that warm air can contain more water vapor.

The $^{7}Be/^{210}Pb$ peaks are in some cases coinciding with ozone peaks but local ^{7}Be activity peaks are found to be more frequently coinciding with local ozone maxima. Thus the radioisotope data are consistent with the hypothesis that maximum ozone values are frequently reached in situations where there is a combined impact of entrainment of ozone rich air brought down by subsidence (high ^{7}Be activities) and stagnant atmospheric conditions (high ^{210}Pb activities), favoring ozone formation in the boundary layer. In such conditions, ^{7}Be activities will be high, but not the $^{7}Be/^{210}Pb$ ratio. This is illustrated in Figure 1 for the year 2011, but a similar picture is seen during other years, where observations are available (see Supplement Figs S1-S4).

As reported in the literature, stratospheric ozone intrusions in the Mediterranean occur most frequently from March to July (Beekmann et al., 1994; Monks et al., 2000). An ozone increase is generally observed during spring months, associated with the increase in solar radiation and photochemistry (Monks et al., 2000) but also from tropospheric downward transport (Kalabokas et al., 2017) and thus some caution would be needed in the data-analysis as the increase in O_3, ^{7}Be and $^{7}Be/^{210}Pb$ at the same time is not necessarily related to the impact of stratospheric intrusions. However, the occurrence of $^{7}Be/^{210}Pb$ peaks above the baseline level can be interpreted as evidence of deep atmospheric subsidence followed by entrainment into the boundary layer. The episodes in May and June 2011 with high ozone and $^{7}Be/^{210}Pb$ values (Fig 1) appears to be due to such deep tropospheric subsidence as indicated by synoptic meteorological maps, back-trajectories and IASI satellite data. Such specific episodes will be examined in detail later in the following section.

Table 2 shows the average monthly values for spring, summer and autumn (March to October) of ^{7}Be, $^{7}Be/^{210}Pb$ and O_3 (12-18 afternoon mean) for all of the years 2006-2015 where measurements were available. The ^{7}Be concentrations show maximum monthly values in June-July, same as ozone, while the $^{7}Be/^{210}Pb$ ratio has its peak values in the April – June period which would indicate enhanced deep tropospheric subsidence during this period of the year.

3.2 Investigation of the main synoptic characteristics during the May- July ozone episodes

As the May-July period seems to be the most important one regarding the influence of subsidence on ozone concentrations, as a next step selected high ^{7}Be and ozone weekly episodes during the May-July period will be examined. More than ten ^{7}Be - ozone weekly episodes were identified in the whole time series and the three most characteristic of them, for what concerns signs of tropospheric subsidence as observed in the meteorological and air pollution measurements (high ^{7}Be and O_3 concentrations combined with positive omega and dry air masses), will be presented in the following paragraphs. The selected episodes were: 3-10 May 2011, 23-29 May 2012 and 28 June – 04 July 2011. The episodes discussed here are not Foehn events.

3.2.1 May 2011 ozone episode

In Figure 2 the composite NOAA/ESRL weather maps of geopotential height, wind speed, omega vertical velocity and specific humidity for the episode of 3-10 May 2011 (as well as the corresponding 3-10 May climatology charts) at JRC-Ispra are shown. A strong low-pressure system is observed over the Atlantic and a weaker one over Eastern Europe and the Black-Sea while anticyclonic conditions prevail over Western and Central Europe in a vast region extending from Scandinavia to the Mediterranean. In this region strong subsidence is observed as deduced from the charts of omega vertical velocity and specific humidity, indicating large-scale downward movement of dry air masses. Similar subsidence conditions have been already observed during regional spring ozone episodes over the western Mediterranean, especially related to the interface of an anticyclone and a low-pressure system located further east (Kalabokas et al., 2017).

3.2.1 May 2012 ozone episode

The above feature is also clearly seen in the following selected episode of 23-29 May 2012 (Fig. 3), with a much stronger anticyclone established over the North Sea (weekly averages measured at Ispra: ^{7}Be: 7117 μBq m^{-3}, $^{7}Be/^{210}Pb$ ratio: 9.6, Ozone 54.9 ppb). The corresponding IASI and IASI+GOME2 satellite images as well as IASI vertical sections at various latitudes from 60-45º N for ozone concentration at 3 km and 10 km are shown in Figs. 4-6. The IASI+GOME2 maps show enhanced ozone concentrations at 3km over Germany, France and Italy, which seem disconnected from the values at 10 km. In a general way, IASI-GOME2 ozone concentrations at 3km seem more independent from those at 10 km, than are those derived from IASI at the same altitudes, which is expected because the number of degrees of freedom is larger. It appears that the geographical distribution of tropospheric ozone as well as the movement of the high ozone reservoirs at both altitude levels generally follow the synoptic weather patterns (Figs. 3, 4). An extended high ozone area appears to the east of the anticyclone at both tropospheric levels apparently originating from the north, being an extension of the northern polar high-ozone reservoir. It has to be added, that back-trajectories show air masses arriving to the Ispra site from N-NE directions and from higher altitudes, especially on May 25-27 (not shown). This behaviour is usually encountered in the analysis of many spring ozone episodes over the area as the 6-day back-trajectories usually originate from the region of high tropospheric ozone subsidence over central and eastern Europe, thus inducing high ozone background levels of tropospheric origin at the boundary layer and at the surface to which the photochemical ozone build-up might eventually be added.

3.2.1 June – July 2011 ozone episode

The next episode of 28 June – 04 July 2011 seems to be quite representative of early summer ozone episodes over the area and will be examined in more details by taking into account many relevant atmospheric measurements recorded at the JRC-Ispra station. During this episodic event, very high ^{7}Be concentrations as well as $^{7}Be/^{210}Pb$ ratio values have been recorded, which were actually the highest weekly averages for 2011 (Figs. 1a,b). Regarding the weather conditions, it has been mostly sunny throughout the week with some rain on the second day and light north-westerly winds.

The synoptic conditions (Figs 7-8, Fig. S5) show the existence of an extended deep low-pressure system over the Atlantic in the free troposphere (at 500 -700hPa), and also another one over central Europe and the boundary layer (at 700-1000hPa), while high atmospheric pressure prevails over most of the European continent, including Scandinavia. Following the examination of the meteorological charts, it has been observed that massive subsidence occurs over a wide area over the Atlantic and western Europe towards the Mediterranean, including the Alpine region, peaking on 1-3 July. Indeed, specific humidity charts at various pressure levels (Fig. S5) show an extended area of dry air masses over the Atlantic (N. Spain – W. France) at lower latitudes, moving towards the European continent and the Mediterranean following the synoptic flow (Fig. 7). The omega vertical velocity charts show that the descending motion is stronger at higher altitudes but at the same time there is an accumulation of dry air masses over the Atlantic (indicating subsidence), which are displaced according to the above described synoptic pattern. In addition, the tropospheric ozone distribution as measured by the IASI and IASI+GOME2 satellites at 3 and 10 km follows the synoptic patterns (Figs. 9-11). As observed on June 30 and July 1, there is a large zone of enhanced ozone at 10 km, but also at 3 km, descending from the North Sea to the Alps, and which corresponds to a through east of the ridge. During July 2 and 3, the through has developed to a cut-off low located over SE Poland, and so do high ozone values at 10 km.

The above described characteristics related to subsidence are much more pronounced if only 1-2 July is considered, especially the large-scale descending tendency of tropospheric air masses at all pressure levels over Central and Northern Europe, which is subsequently associated with extended tropospheric dryness over these areas (Figs S6-S8).

In addition, according to the back-trajectory plots the subsiding air masses arrive to the JRC-Ispra site from the Atlantic coast and France (Fig. 16). Almost all back-trajectories arrive from the north, where the subsidence area is located (Figs 9-10, Fig. S5) and where a large area of tropospheric ozone appears over Western Europe, apparently originating from the high ozone reservoir located over polar regions as shown in the corresponding IASI and IASI+GOME2 satellite images as well as IASI vertical sections (Figs 9-11).

The hourly air pollution measurements at the JRC-station during the 28 June – 04 July 2011 period show first a period (June 28 and 29) with large ozone (daily maximum more than 80 ppb) and PM_{10} (daily maximum more than 80 $\mu g/m^3$), related to high temperatures (T_{max} 30°C and low wind speed (<1 m/s). Aerosol is of mainly anthropogenic origin as indicated by different nephelometer responses to red, green and blue light. High ozone values during this period are probably due mainly to photochemical build-up from anthropogenic emissions in the Po valley. During the following period with maximum of subsidence (1-2 July), ozone concentrations vary around 55-60 ppb with the diurnal concentration range significantly reduced in comparison to the previous days while at the same time the NO_x concentrations get minimum values (Fig. 13a) as well as the humidity, ^{222}Rn, nephelometer and PM_{10} concentrations (Figs. 13b-d). Please note that the weekly resolution of ^{7}Be measurements does not allow for ascertaining an expected maximum during these two days. During the days following the strongest subsidence period (3-4 July) the nocturnal ozone values are significantly reduced as the tropospheric entrainment has diminished while the ozone destruction by NO chemical titration and dry deposition on the ground reduces substantially the concentrations within the generally shallow nocturnal layer. On the other hand, the mid-day ozone concentrations are slightly increased probably due to in-situ photochemical ozone production, which is added-up to the increased tropospheric ozone background, due to the regional tropospheric subsidence episode occurring during the previous days. Thus, this period shows an interesting suite of days with strong photochemical ozone production and advection of tropospheric ozone to the ground.

In summarizing the above, it has to be mentioned at first that maximum ozone values in the area of the study are expected to be connected primarily with boundary layer ozone photochemistry. The presented analysis shows that regional tropospheric subsidence occurring frequently in the area during May – July might enable easier exceedances of ozone air quality standards, as photochemical ozone build-up is initiated in a clean boundary layer air mass containing already high ozone levels (i.e. 50 ppb), which is common after a regional tropospheric subsidence episode in the area. This phenomenon may explain the spring ozone maximum observed over many stations over the Western and Central part of the European continent including the Western Mediterranean. In relation to the above, it has been mentioned in the introduction that a similar phenomenon is observed in the Eastern Mediterranean, maximizing though later in summer (July – August).

4 Conclusion

The present study focuses on the influence of tropospheric subsidence on surface ozone concentrations, especially during spring and summer months over the western Mediterranean area. It is a much more detailed extension of a previous study (Kalabokas et al., 2017) where some troposheric mechanisms related with regional ozone episodes especially linked with large-scale subsidence were examined. This study is focused on a more detailed analysis of the suggested mechanisms, based on the measurements of ^{7}Be , ^{210}Pb as well as a very large variety of meteorological and air pollution parameters collected at the JRC-Ispra station, which is considered to be one of the most well-equipped measuring sites in Europe. High boundary layer concentrations of ^{7}Be are used as indicator of the influence of free tropospheric air, in which ^{7}Be activity is large due a cosmogenic source. ^{210}Pb activity is an indicator of accumulation of surface emissions and their reaction products and it reaches its highest levels during periods with stagnant air conditions. Radioisotope and humidity data from the Ispra station show that local peaks in $^{7}Be/^{210}Pb$ frequently are coinciding with local minima in specific humidity, consistent with the hypothesis that these peaks are found in situations with a strong impact of free tropospheric influence to the boundary layer. Comparison with ozone measurements shows that these peaks in some occasions coincide with ozone peaks but more frequently ozone peaks coincide with peaks in ^{7}Be activity. This observation was interpreted as a result of the fact that the highest ozone concentrations frequently are found in situations with a combined impact of entrainment of ozone rich free tropospheric air and local formation in the boundary layer. The conclusions derived from the analysis of specific episodes were in accordance with this interpretation: The main characteristics of the frequently occurring spring episodes where both ^{7}Be and ozone reach maxima at the Ispra station were found to be the following:

* Anticyclonic stagnant conditions over parts of the European continent and the western Mediterranean,
* Strong winds at the periphery of the anticyclone associated with a deep low-pressure system located to the North and a weaker one located to the East. A common feature is that Ispra is located at the eastern edge of a ridge system at 700 hPa level and at the same time a trough is located eastwards,
* Very extended areas of positive vertical velocity, omega (downward movement), observed over eastern, central and western Europe, depending on the locations and the relative strength of the high and low pressure systems, at all pressure levels and associated with dry conditions (low specific humidity), indicating subsidence.

At the same time, the IASI satellite images show important ozone reservoirs in the upper and lower troposphere, which are generally delimitated by the meteorological systems and follow their movement while large areas of enhanced tropospheric ozone appear over the region of subsidence, usually originating from the tropospheric ozone reservoirs associated with the low-pressure systems. These results consolidate the findings of the first phase of this study on spring ozone episodes in the western Mediterranean (Kalabokas et al., ACP, 2017), and extend them over a full year and a longer time period. The characteristics described above are also encountered during some summer episodes (in June-July) but the conditions generally observed in summer episodes are more related to local photochemical ozone production in the boundary layer while tropospheric subsidence is weaker and more concentrated over the Eastern Mediterranean.

The most important new findings of this paper are:

a) The examination of the Ispra station hourly measurements during subsidence episodes shows that the local pollution parameters (e.g. NO_x, ^{222}Rn, nephelometer, PM_{10}) tend to have low values (as compared to those observed during periods of anthropogenic pollution), while the ozone levels usually reach values around 45-60 ppb during the afternoon hours but show significantly higher values than the average during the night and morning hours. This is a clear sign of tropospheric entrainment to the boundary layer.

b) During high ^{7}Be and high ozone episodes, the highest ozone values exceeding the standards usually occur within the following 2-3 days after the maximum of regional tropospheric subsidence. The increase in ozone concentrations usually occurs under the influence of favourable meteorological conditions for photochemical ozone production in the boundary layer, which is added-up on the increased regional background due to tropospheric subsidence and thus occasionally leading to exceedances in ozone air quality standards.

The results of this study might be useful for helping the required improvements in the veracity of global ozone air quality models for which biases have been made evident by several recent studies (Cooper et al., 2014; Parrish et al., 2014; Gaudel et al., 2018; Young et al., 2018). Indeed, these models need to accurately represent complex vertical transport processes affecting the surface ozone budget.

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Author contributions

PK, NRJ and JH prepared the manuscript with contributions from all co-authors to the manuscript/data-evaluation. MR provided ^{7}Be and ^{210}Pb data. NRJ and JH provided ozone and NO_x data. PK provided synoptic meteorological maps and back-trajectories. GF, GD, ME and MB provided the IASI satellite ozone measurements.

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Table 1. Instruments and measurements at JRC-Ispra site.

Variable	Instrument	Technique	Unit	Resolution	Reference
Ozone	Thermo 49	UV	ppb	Hourly and 6-hour daily average	Putaud et al., 2017
^{7}Be, ^{210}Pb	GEM series EG&G Ortec	Filter sampling, HpGe coaxial detectors	µBq m^{-3}	Weekly average	Jensen et al., 2017
^{222}Rn	ANSTO (custom build)	Filter sampling, ANSTO detector	µBq m^{-3}	Hourly average	Zahorowski et al., 2004 Putaud et al., 2017
NO_x (NO,NO_2)	Thermo 42/42iTL	Chemiluminesc.	ppb	Hourly average	Villena et al., 2012 Putaud et al., 2017
Particle light scattering	Integrating Nephelometer TSI 3563	Light-integration	km^{-1}	Hourly average	Putaud et al., 2017
PM_{10}	FDMS TEOM	TEOM	µg m^{-3}	Hourly average	Putaud et al., 2017
Relative Humidity[*)]	WXT510	Capative thin-film sensor	ppm	Hourly and 6-hour average	Putaud et al., 2017

[*)] Other meterological variables measured: temperature, pressure, precipitation, wind speed and wind direction (Putaud et al., 2017).

Table 2: Monthly averages of ^{7}Be, ^{210}Pb, ozone (12:00 – 18:00) concentrations and ^{7}Be/ ^{210}Pb ratio during the March – October period (2006-2015).

Month	^{7}Be (μBq m^{-3})	^{210}Pb (μBq m^{-3})	^{7}Be/^{210}Pb ratio	O_3 12-18 (ppb)
3	4031.2	875.3	6.1	25.0
4	4737.1	835.4	7.1	34.9
5	4741.0	726.7	7.3	40.2
6	4863.8	794.4	7.0	52.6
7	5824.5	1049.7	6.6	52.5
8	5272.3	1114.8	5.7	36.5
9	4490.8	1090.8	4.7	25.4
10	4249.3	1609.7	3.6	16.3

FIGURE CAPTIONS

Figure 1. (a): Weekly averages for ozone 12:00-18:00 (ppb, red) and ^{7}Be (mBq m^{-3}, blue) at the JRC-Ispra station for 2011. The authors acknowledge the use of a similar figure in ref. Jensen et al., 2017 (see the 'acknowledgements' section for more details).
(b): Weekly averages for ozone 12:00-18:00 (ppb, red) and ^{7}Be/^{210}Pb ratio (black) at the JRC-Ispra station for 2011. The authors acknowledge the use of a similar figure in ref. Jensen et al., 2017 (see the 'acknowledgements' section for more details).
(c): Weekly averages of the ^{7}Be concentrations (mBq m^{-3}, red) and specific humidity 12:00-18:00 (ppmV, blue) at the JRC-Ispra station for 2011.
(d): Weekly averages of the ^{7}Be /^{210}Pb ratio and specific humidity 12:00-18:00 (ppmV, blue)at the JRC-Ispra station for 2011.

Figure 2: Composite NOAA/ESRL weather maps of geopotential height, vector wind speed, omega vertical velocity and specific humidity for 3-10 May climatology (left column) and for the episode of 3-10 May 2011 at JRC-Ispra, Italy (right column).

Figure 3: Composite NOAA/ESRL weather maps of geopotential height, vector wind speed, omega vertical velocity and specific humidity for 23-29 May climatology (left column) and for the episode of 23-29 May 2012 at JRC-Ispra, Italy (right column).

Fig. 4: IASI satellite images for ozone concentration at 3 km (left column) and 10 km (right column) for the episode of 23-29 May 2012. Starting from the top: 23-29 May, 23-24 May, 25-27 May, 28-29 May.

Fig. 5: IASI vertical sections at various latitudes from 65° N (upper panel) to 45 ° N (lower panel) on May 23-24, 2012 (left column) and on May 25-27, 2012 (right column).

Fig. 6: IASI +GOME2 satellite images for ozone concentration at 3 km (left column) and 10 km (right column) for the episode of 23-29 May 2012. Starting from the top: 23-29 May, 23-24 May, 25-27 May, 28-29 May.

Figure 7: Composite NOAA/ESRL weather maps of geopotential height, vector wind speed, omega vertical velocity and specific humidity for 28 June-04 July 2011 climatology (left column) and for the episode of 28 June-04 July 2011 at JRC-Ispra, Italy (right column).

Fig. 8: Composite charts for specific humidity anomaly at 850 hPa (left column) and at 1000 hPa (right column), 5-days, 3-days, 2-days before and during the episode of 28 June-04 July 2011 (lower panels).

Fig. 9: IASI satellite images of ozone concentration at 3 km (left column) and 10 km (right column) for the episode of 28 June – 04 July 2011. Starting from the top: 28 June – 04 July , 30 June – 01 July, 02 July – 03 July.

Fig. 10: IASI vertical sections at various latitudes from 65 ° N (upper panel) to 45 N (lower panel) on June 30-July 1, 2011 (left column) and on July 2-3, 2011 (right column).

Fig. 11: IASI+GOME2 satellite images of ozone concentration at 3 km (left column) and 10 km (right column) for the episode of 28 June – 04 July 2011. Starting from the top: 28 June – 04 July, 30 June – 01 July, 02 July – 03 July.

Fig. 12: 6-day HYSPLIT back-trajectories arriving at the JRC-Ispra station during the episode of 28 June-04 July 2011.

Fig. 13: Air pollution and meteorological measurements for 28 June-04 July 2011 at JRC-Ispra, Italy.

(a): Hourly ozone (red) and nitrogen dioxide (brown) mixing ratios. (b): Hourly Relative Humidity (blue) and temperature (red) measurements. (c): Hourly ^{222}Rn concentrations (in Bq m^{-3}, black) and Wind Speed (in m s^{-1}, blue) for 28 June-04 July 2011 at JRC-Ispra, Italy. (d): Hourly PM_{10} (in black, µg*m^{-}3) and Nephelometer (in blue, green and red) measurements (in m^{-1}).

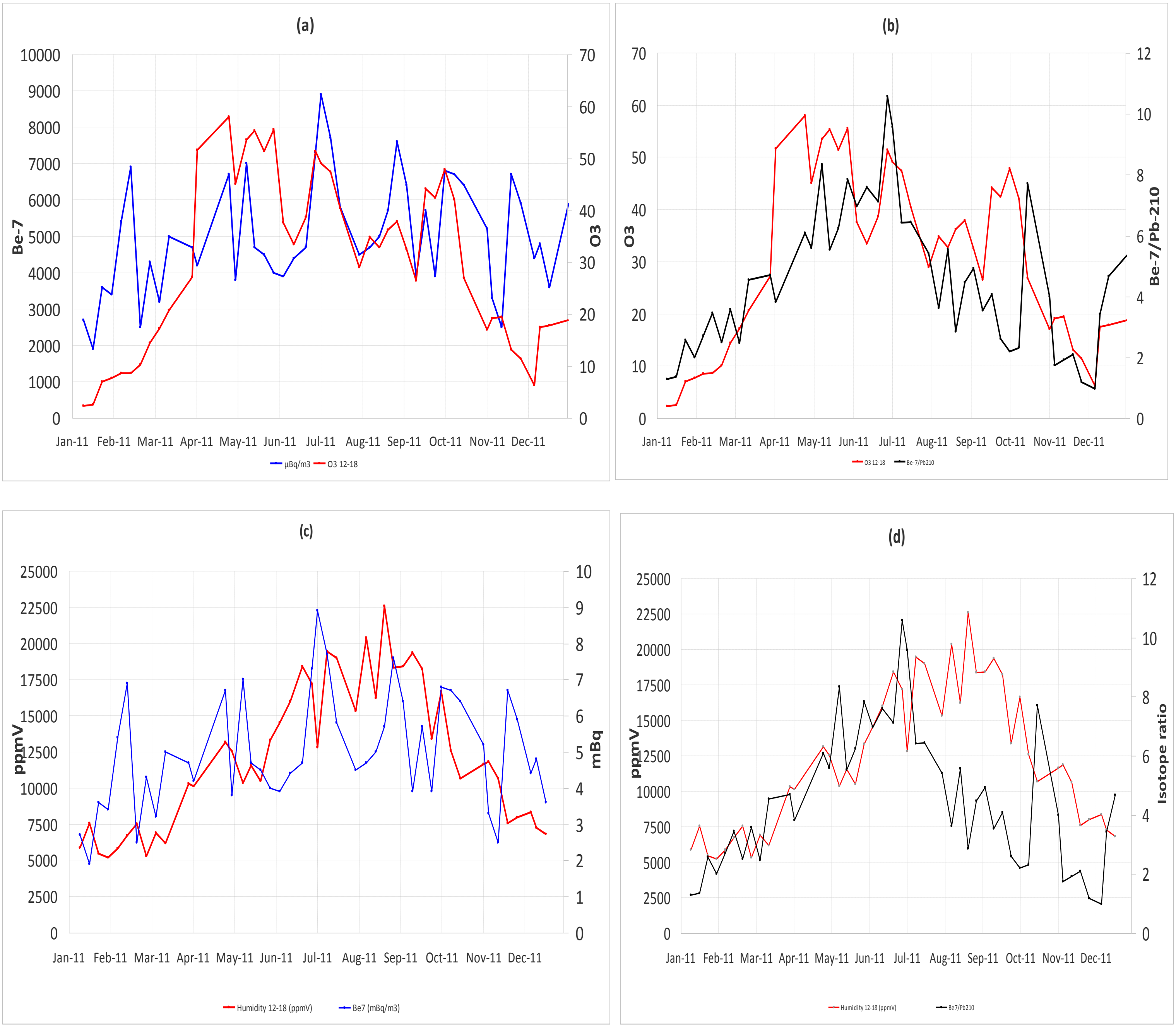

(a)
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
Be-7
70
60
50
40
30
20
10
0
O3
Jan-11
Feb-11
Mar-11
Apr-11
May-11
Jun-11
Jul-11
Aug-11
Sep-11
Oct-11
Nov-11
Dec-11
µBq/m3
O3 12-18
(b)
70
60
50
40
30
20
10
0
O3
12
10
8
6
4
2
0
Be-7/Pb-210
Jan-11
Feb-11
Mar-11
Apr-11
May-11
Jun-11
Jul-11
Aug-11
Sep-11
Oct-11
Nov-11
Dec-11
(c)
25000
22500
20000
17500
15000
12500
10000
7500
5000
2500
0
ppmV
10
9
8
7
6
5
4
3
2
1
0
mBq
Jan-11
Feb-11
Mar-11
Apr-11
May-11
Jun-11
Jul-11
Aug-11
Sep-11
Oct-11
Nov-11
Dec-11
Humidity 12-18 (ppmV)
Be7 (mBq/m3)
(d)
25000
22500
20000
17500
15000
12500
10000
7500
5000
2500
0
ppmV
12
10
8
6
4
2
0
Isotope ratio
Jan-11
Feb-11
Mar-11
Apr-11
May-11
Jun-11
Jul-11
Aug-11
Sep-11
Oct-11
Nov-11
Dec-11

Figure 1:

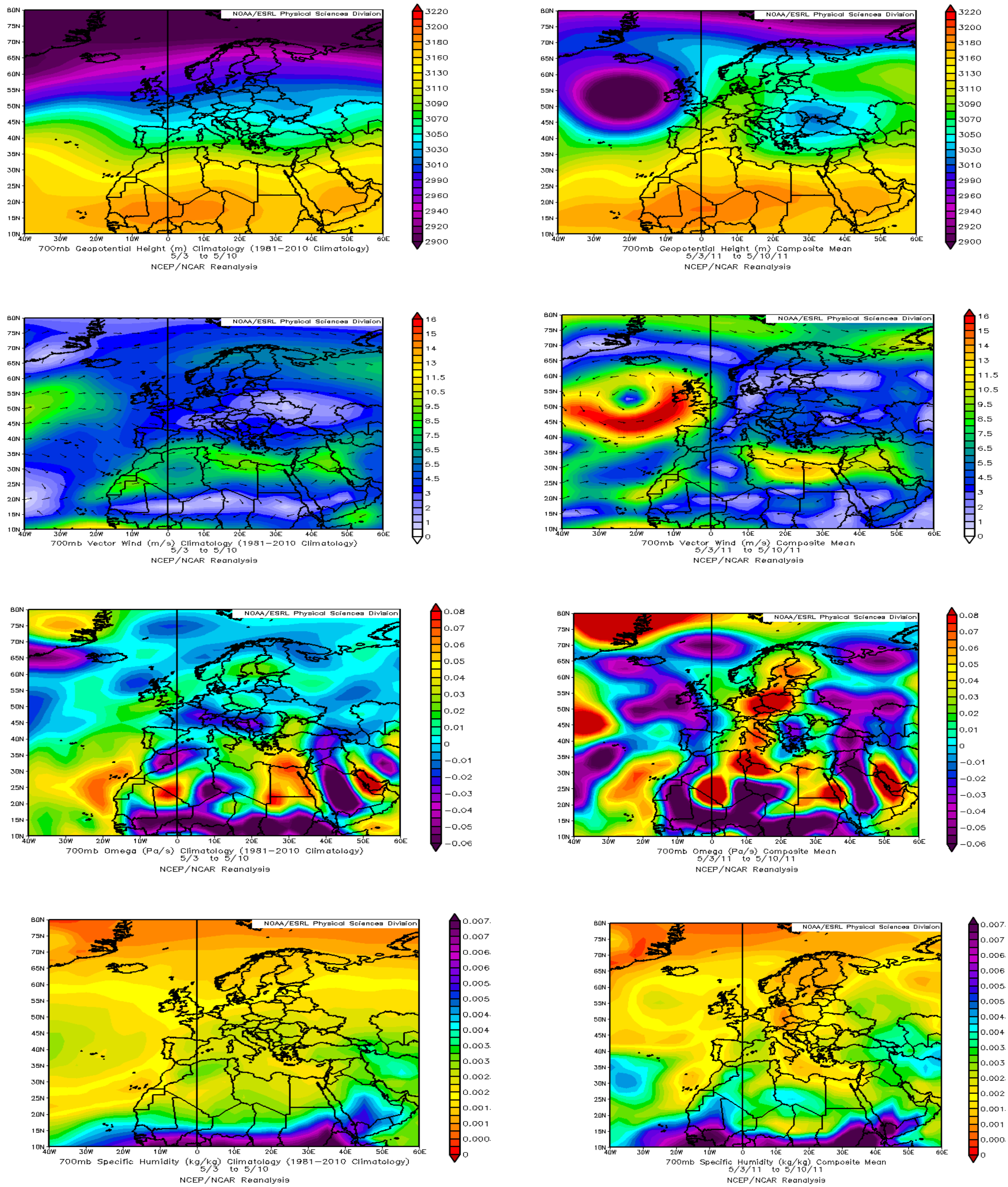
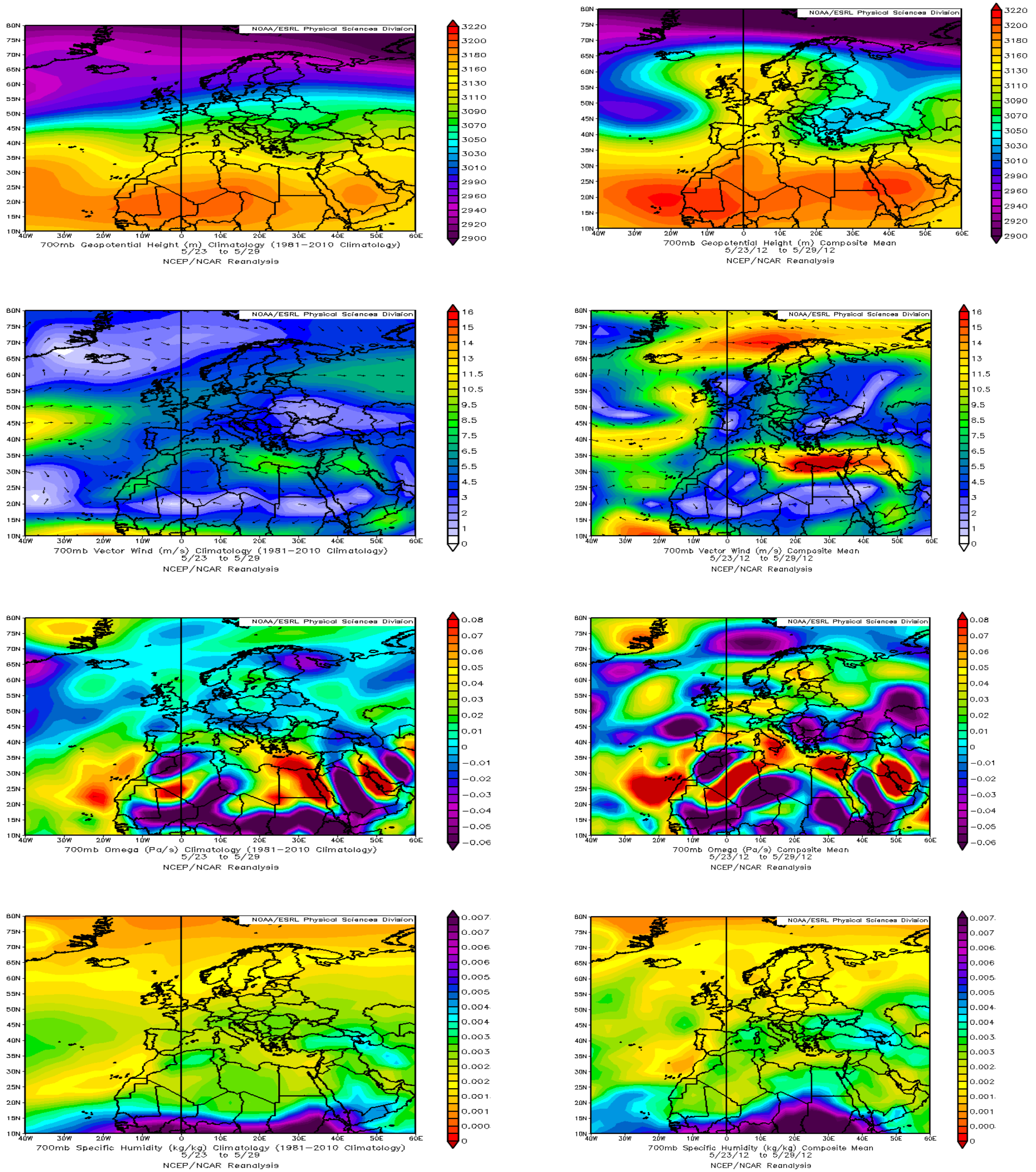

NOAA/ESRL Physical Sciences Division
700mb Geopotential Height (m) Climatology (1981–2010 Climatology)
5/3 to 5/10
NCEP/NCAR Reanalysis
700mb Geopotential Height (m) Composite Mean
5/3/11 to 5/10/11
NCEP/NCAR Reanalysis
700mb Vector Wind (m/s) Climatology (1981–2010 Climatology)
5/3 to 5/10
NCEP/NCAR Reanalysis
700mb Vector Wind (m/s) Composite Mean
5/3/11 to 5/10/11
NCEP/NCAR Reanalysis
700mb Omega (Pa/s) Climatology (1981–2010 Climatology)
5/3 to 5/10
NCEP/NCAR Reanalysis
700mb Omega (Pa/s) Composite Mean
5/3/11 to 5/10/11
NCEP/NCAR Reanalysis
700mb Specific Humidity (kg/kg) Climatology (1981–2010 Climatology)
5/3 to 5/10
NCEP/NCAR Reanalysis
700mb Specific Humidity (kg/kg) Composite Mean
5/3/11 to 5/10/11
NCEP/NCAR Reanalysis

Figure 2:



NOAA/ESRL Physical Sciences Division
700mb Geopotential Height (m) Climatology (1981–2010 Climatology)
5/23 to 5/29
NCEP/NCAR Reanalysis
700mb Geopotential Height (m) Composite Mean
5/23/12 to 5/29/12
NCEP/NCAR Reanalysis
700mb Vector Wind (m/s) Climatology (1981–2010 Climatology)
5/23 to 5/29
NCEP/NCAR Reanalysis
700mb Vector Wind (m/s) Composite Mean
5/23/12 to 5/29/12
NCEP/NCAR Reanalysis
700mb Omega (Pa/s) Climatology (1981–2010 Climatology)
5/23 to 5/29
NCEP/NCAR Reanalysis
700mb Omega (Pa/s) Composite Mean
5/23/12 to 5/29/12
NCEP/NCAR Reanalysis
700mb Specific Humidity (kg/kg) Climatology (1981–2010 Climatology)
5/23 to 5/29
NCEP/NCAR Reanalysis
700mb Specific Humidity (kg/kg) Composite Mean
5/23/12 to 5/29/12
NCEP/NCAR Reanalysis

Figure 3:

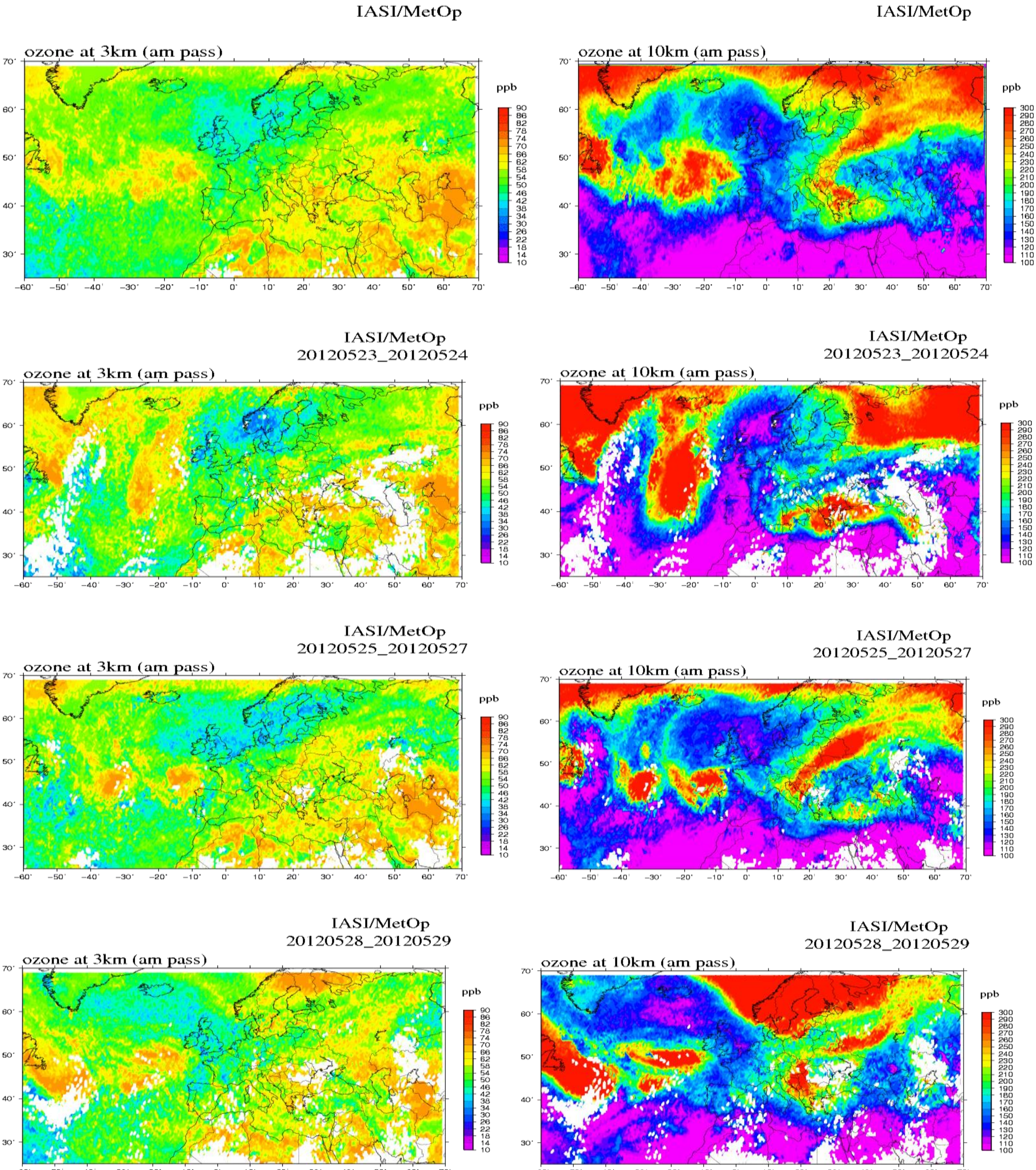

IASI/MetOp
ozone at 3km (am pass)
IASI/MetOp
ozone at 10km (am pass)
IASI/MetOp
20120523_20120524
ozone at 3km (am pass)
IASI/MetOp
20120523_20120524
ozone at 10km (am pass)
IASI/MetOp
20120525_20120527
ozone at 3km (am pass)
IASI/MetOp
20120525_20120527
ozone at 10km (am pass)
IASI/MetOp
20120528_20120529
ozone at 3km (am pass)
IASI/MetOp
20120528_20120529
ozone at 10km (am pass)
ppb

Fig. 4:

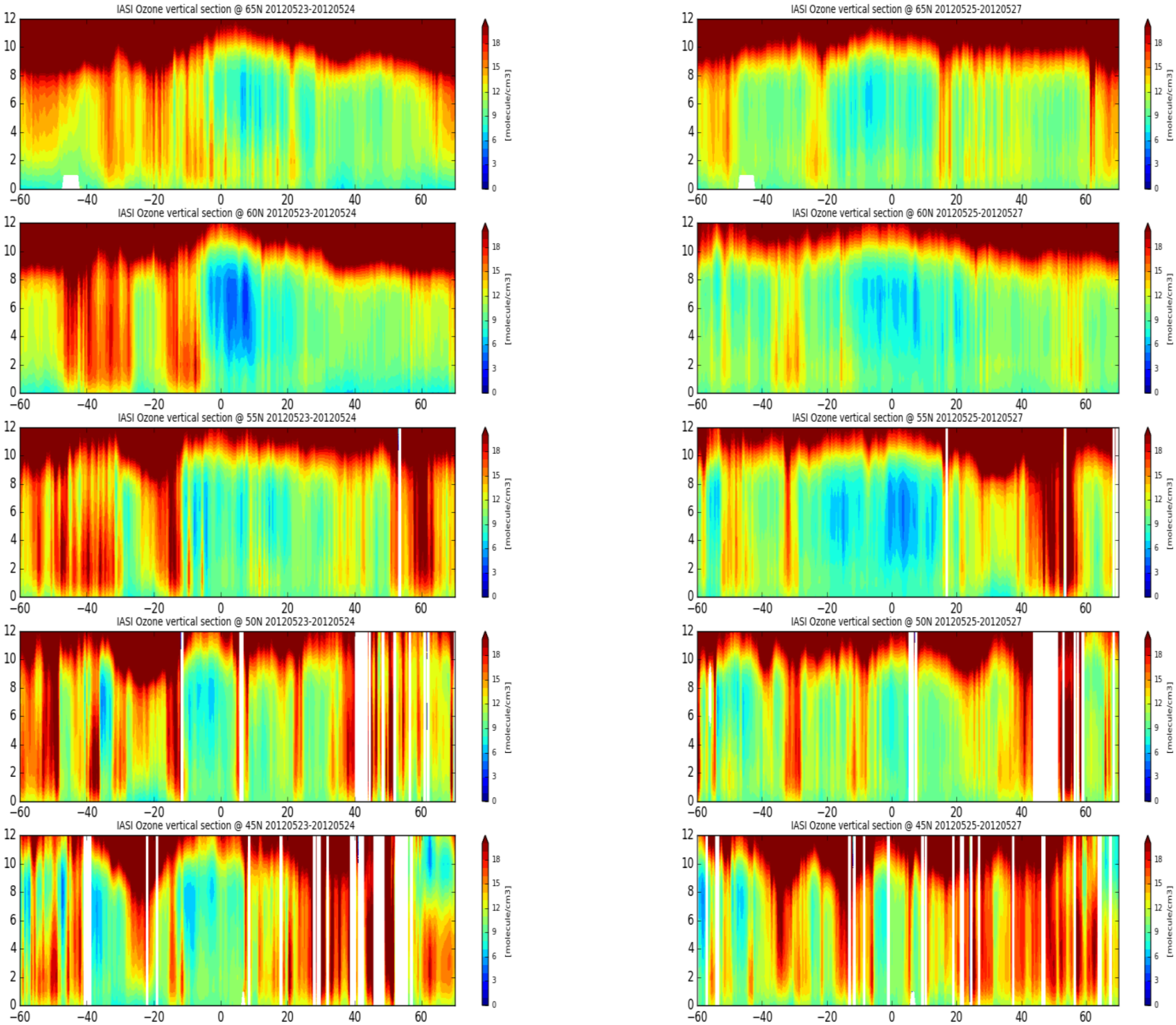

IASI Ozone vertical section @ 65N 20120523-20120524
IASI Ozone vertical section @ 65N 20120525-20120527
IASI Ozone vertical section @ 60N 20120523-20120524
IASI Ozone vertical section @ 60N 20120525-20120527
IASI Ozone vertical section @ 55N 20120523-20120524
IASI Ozone vertical section @ 55N 20120525-20120527
IASI Ozone vertical section @ 50N 20120523-20120524
IASI Ozone vertical section @ 50N 20120525-20120527
IASI Ozone vertical section @ 45N 20120523-20120524
IASI Ozone vertical section @ 45N 20120525-20120527
[molecule/cm3]

Fig. 5:

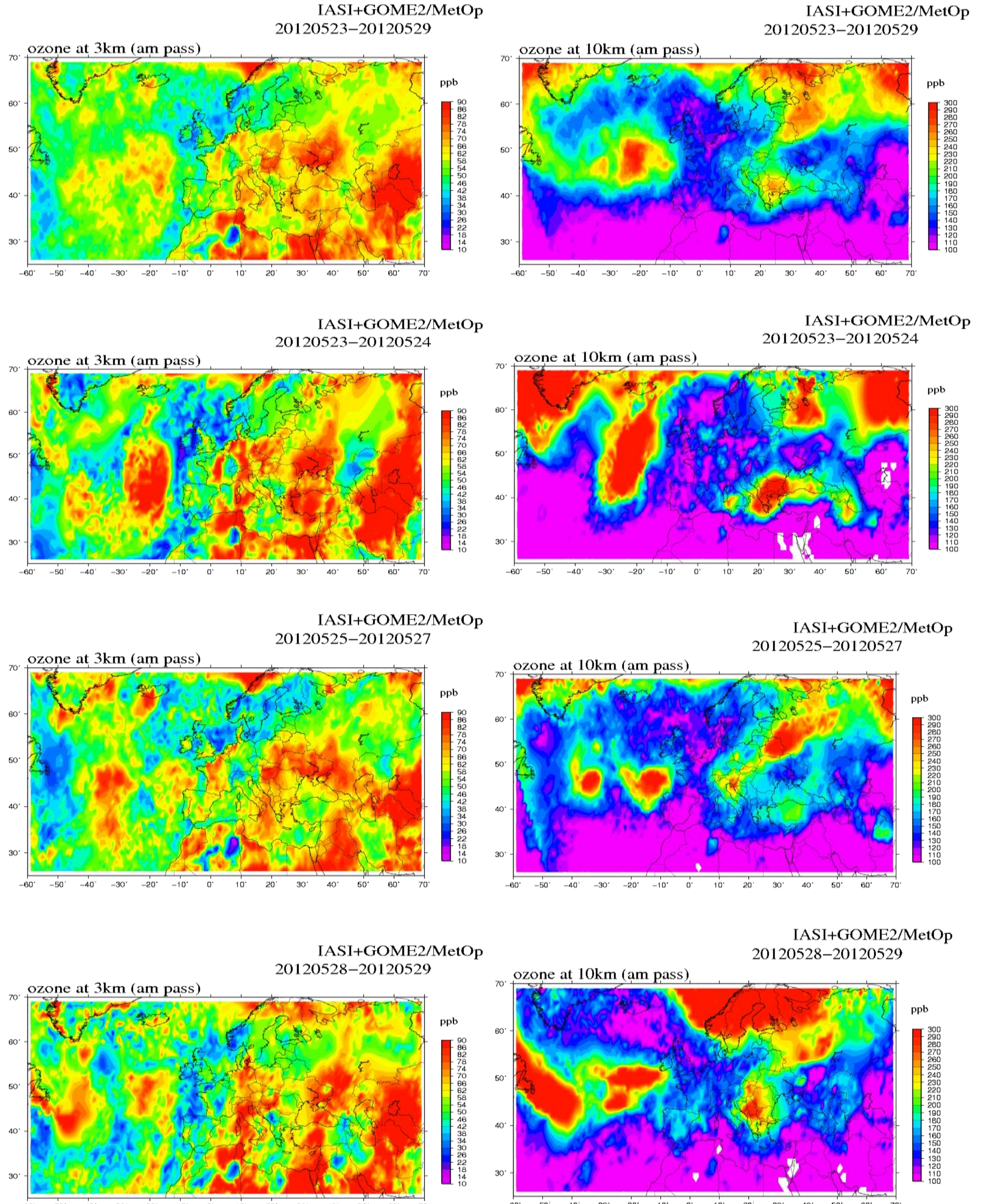

IASI+GOME2/MetOp
20120523−20120529
ozone at 3km (am pass)
IASI+GOME2/MetOp
20120523−20120529
ozone at 10km (am pass)
IASI+GOME2/MetOp
20120523−20120524
ozone at 3km (am pass)
IASI+GOME2/MetOp
20120523−20120524
ozone at 10km (am pass)
IASI+GOME2/MetOp
20120525−20120527
ozone at 3km (am pass)
IASI+GOME2/MetOp
20120525−20120527
ozone at 10km (am pass)
IASI+GOME2/MetOp
20120528−20120529
ozone at 3km (am pass)
IASI+GOME2/MetOp
20120528−20120529
ozone at 10km (am pass)
ppb

Fig. 6:

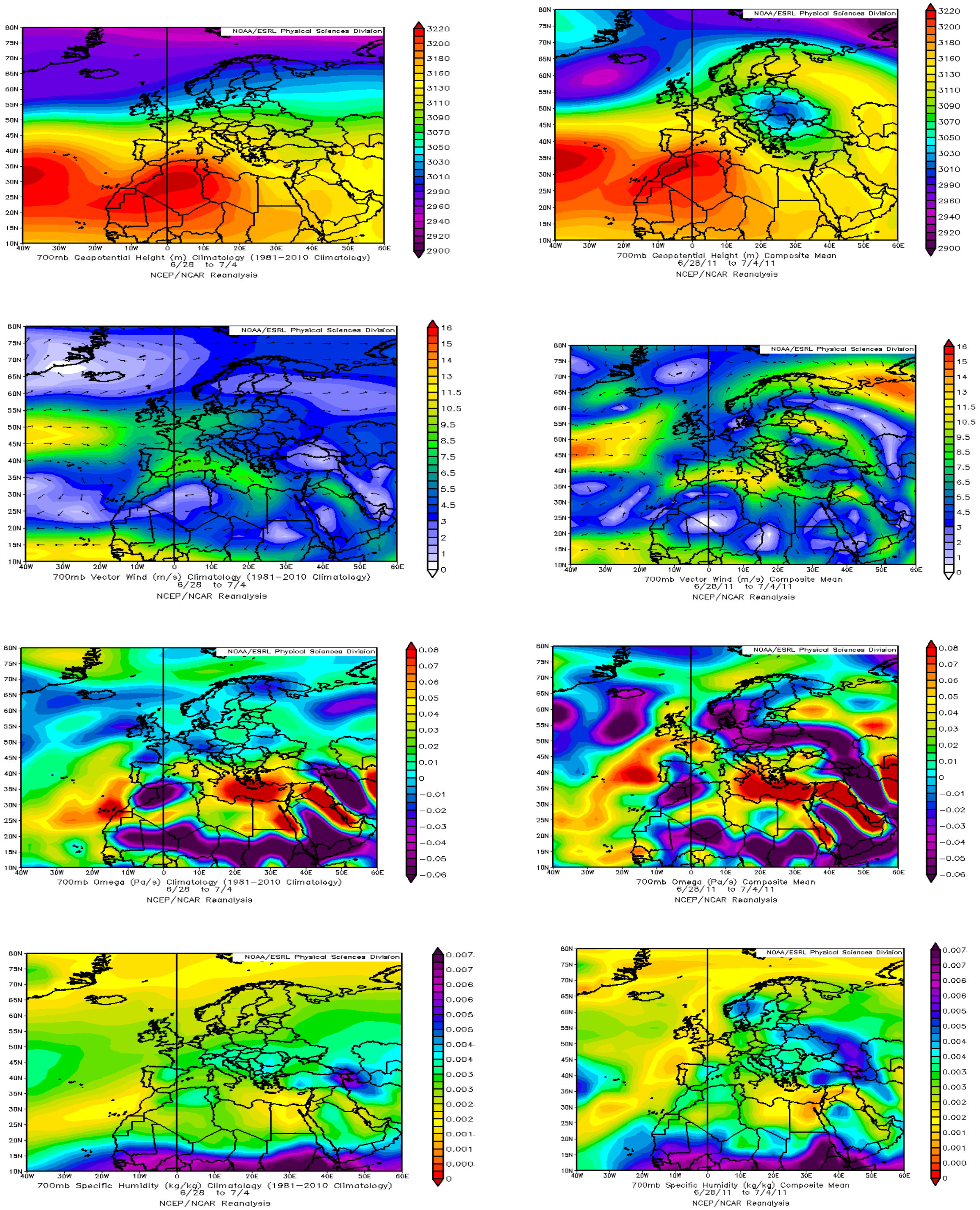

NOAA/ESRL Physical Sciences Division
700mb Geopotential Height (m) Climatology (1981-2010 Climatology)
6/28 to 7/4
NCEP/NCAR Reanalysis
700mb Geopotential Height (m) Composite Mean
6/28/11 to 7/4/11
NCEP/NCAR Reanalysis
700mb Vector Wind (m/s) Climatology (1981-2010 Climatology)
6/28 to 7/4
NCEP/NCAR Reanalysis
700mb Vector Wind (m/s) Composite Mean
6/28/11 to 7/4/11
NCEP/NCAR Reanalysis
700mb Omega (Pa/s) Climatology (1981-2010 Climatology)
6/28 to 7/4
NCEP/NCAR Reanalysis
700mb Omega (Pa/s) Composite Mean
6/28/11 to 7/4/11
NCEP/NCAR Reanalysis
700mb Specific Humidity (kg/kg) Climatology (1981-2010 Climatology)
6/28 to 7/4
NCEP/NCAR Reanalysis
700mb Specific Humidity (kg/kg) Composite Mean
6/28/11 to 7/4/11
NCEP/NCAR Reanalysis

Figure 7:

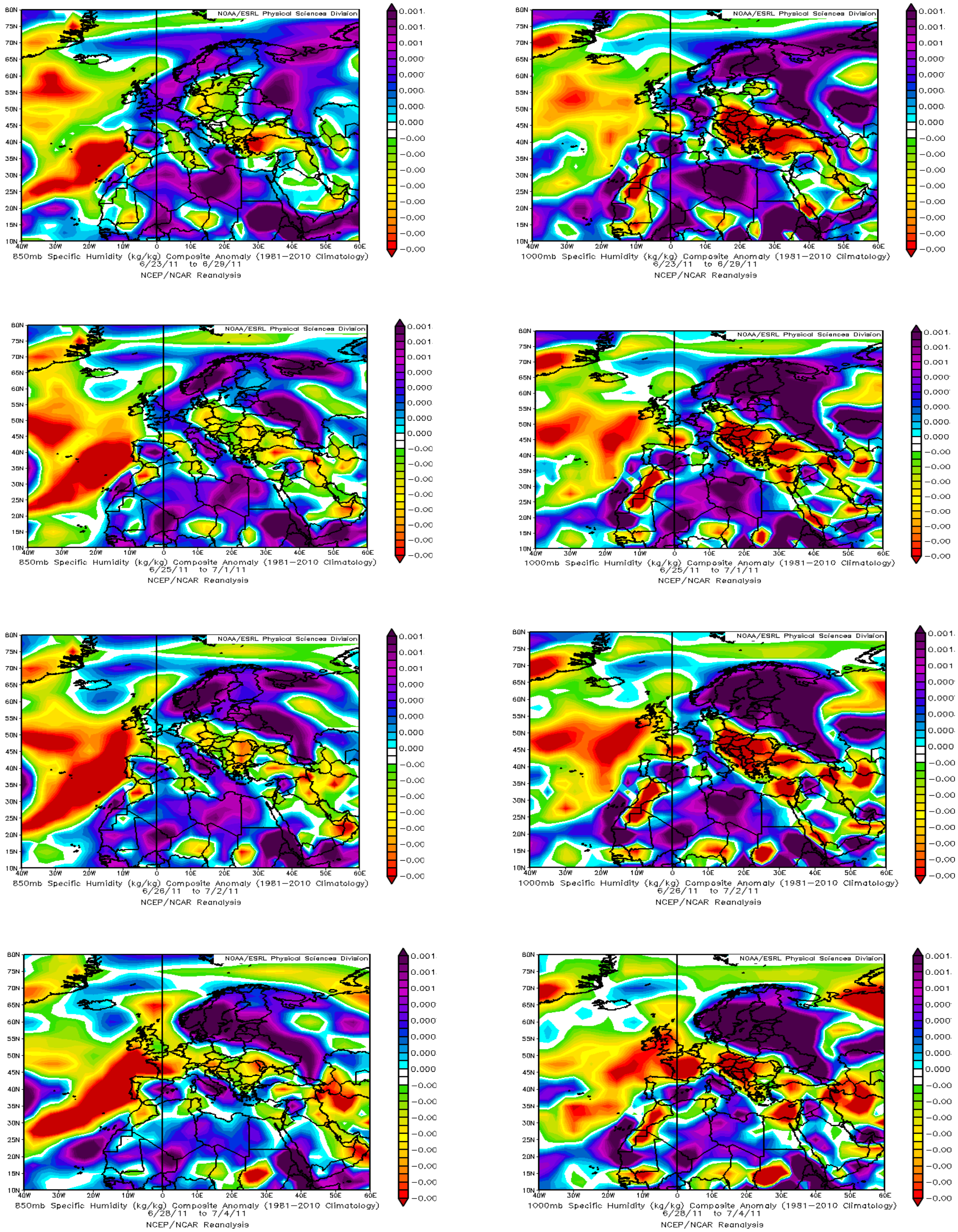
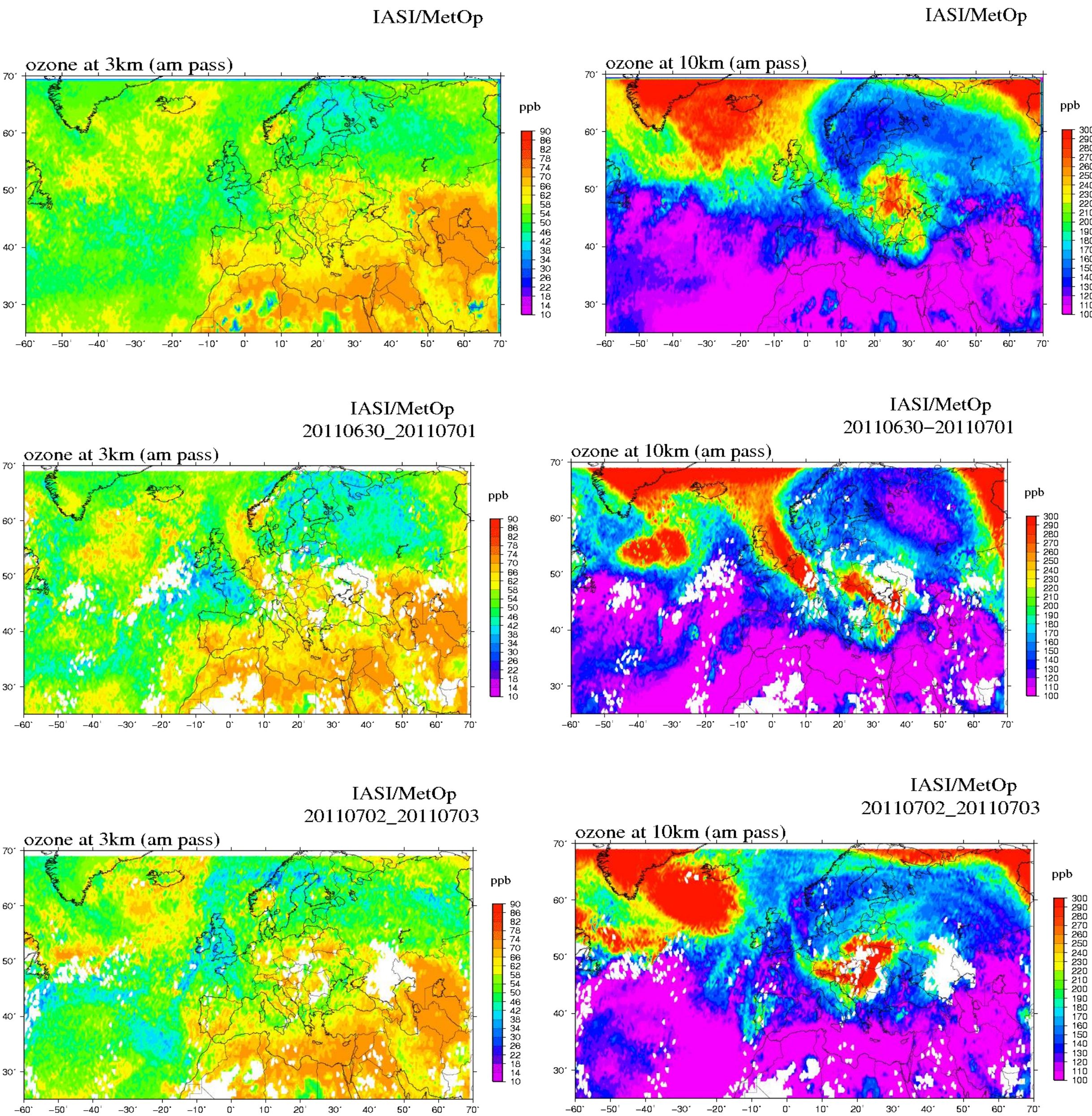
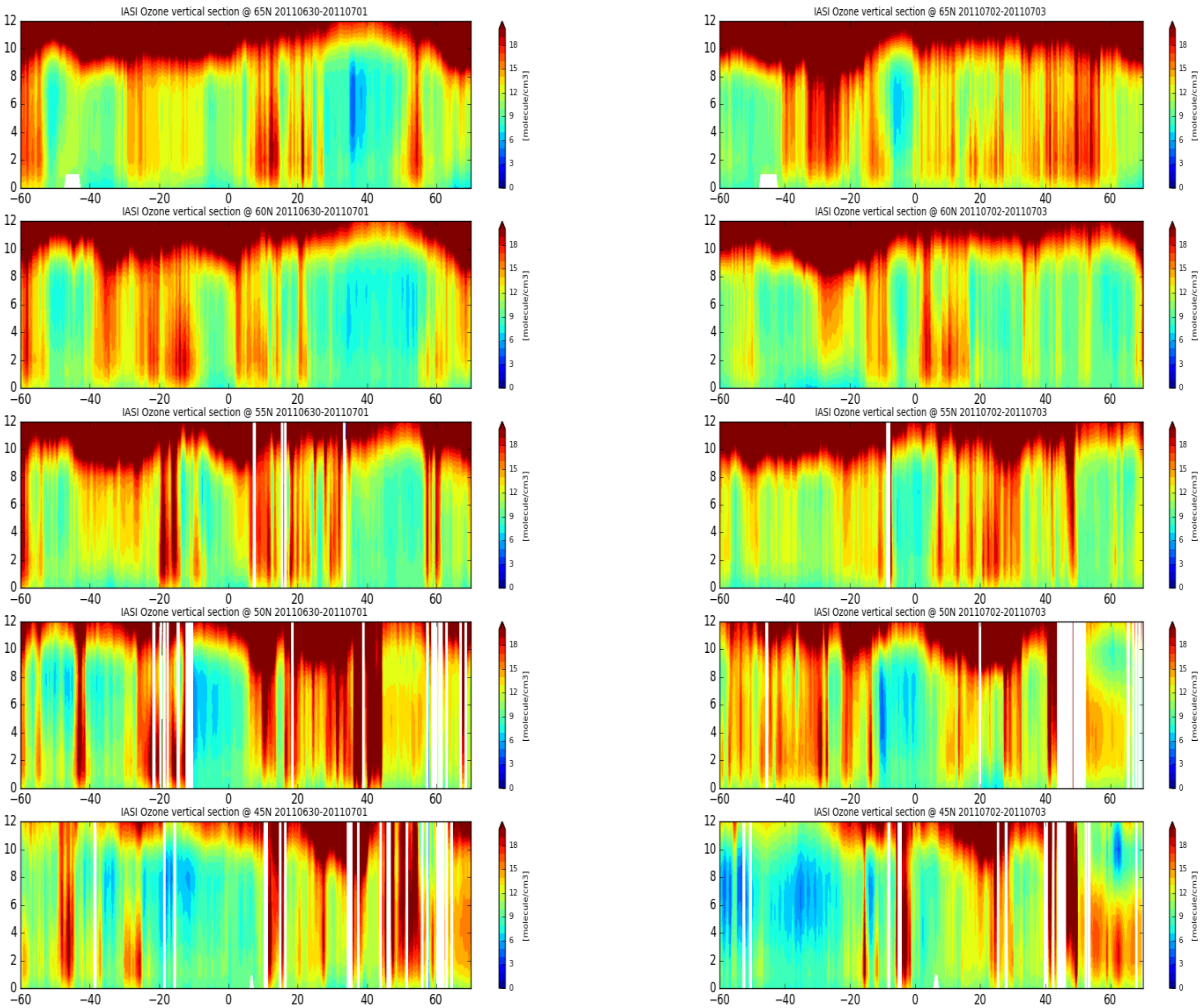

NOAA/ESRL Physical Sciences Division
850mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/23/11 to 6/29/11
NCEP/NCAR Reanalysis
1000mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/23/11 to 6/29/11
NCEP/NCAR Reanalysis
850mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/25/11 to 7/1/11
NCEP/NCAR Reanalysis
1000mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/25/11 to 7/1/11
NCEP/NCAR Reanalysis
850mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/26/11 to 7/2/11
NCEP/NCAR Reanalysis
1000mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/26/11 to 7/2/11
NCEP/NCAR Reanalysis
850mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/28/11 to 7/4/11
NCEP/NCAR Reanalysis
1000mb Specific Humidity (kg/kg) Composite Anomaly (1981–2010 Climatology)
6/28/11 to 7/4/11
NCEP/NCAR Reanalysis

Fig. 8:



IASI/MetOp
ozone at 3km (am pass)
ppb
IASI/MetOp
ozone at 10km (am pass)
ppb
IASI/MetOp
20110630_20110701
ozone at 3km (am pass)
ppb
IASI/MetOp
20110630–20110701
ozone at 10km (am pass)
ppb
IASI/MetOp
20110702_20110703
ozone at 3km (am pass)
ppb
IASI/MetOp
20110702_20110703
ozone at 10km (am pass)
ppb

Fig. 9:


IASI Ozone vertical section @ 65N 20110630-20110701
IASI Ozone vertical section @ 65N 20110702-20110703
IASI Ozone vertical section @ 60N 20110630-20110701
IASI Ozone vertical section @ 60N 20110702-20110703
IASI Ozone vertical section @ 55N 20110630-20110701
IASI Ozone vertical section @ 55N 20110702-20110703
IASI Ozone vertical section @ 50N 20110630-20110701
IASI Ozone vertical section @ 50N 20110702-20110703
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IASI Ozone vertical section @ 45N 20110702-20110703
[molecule/cm3]

Fig. 10:

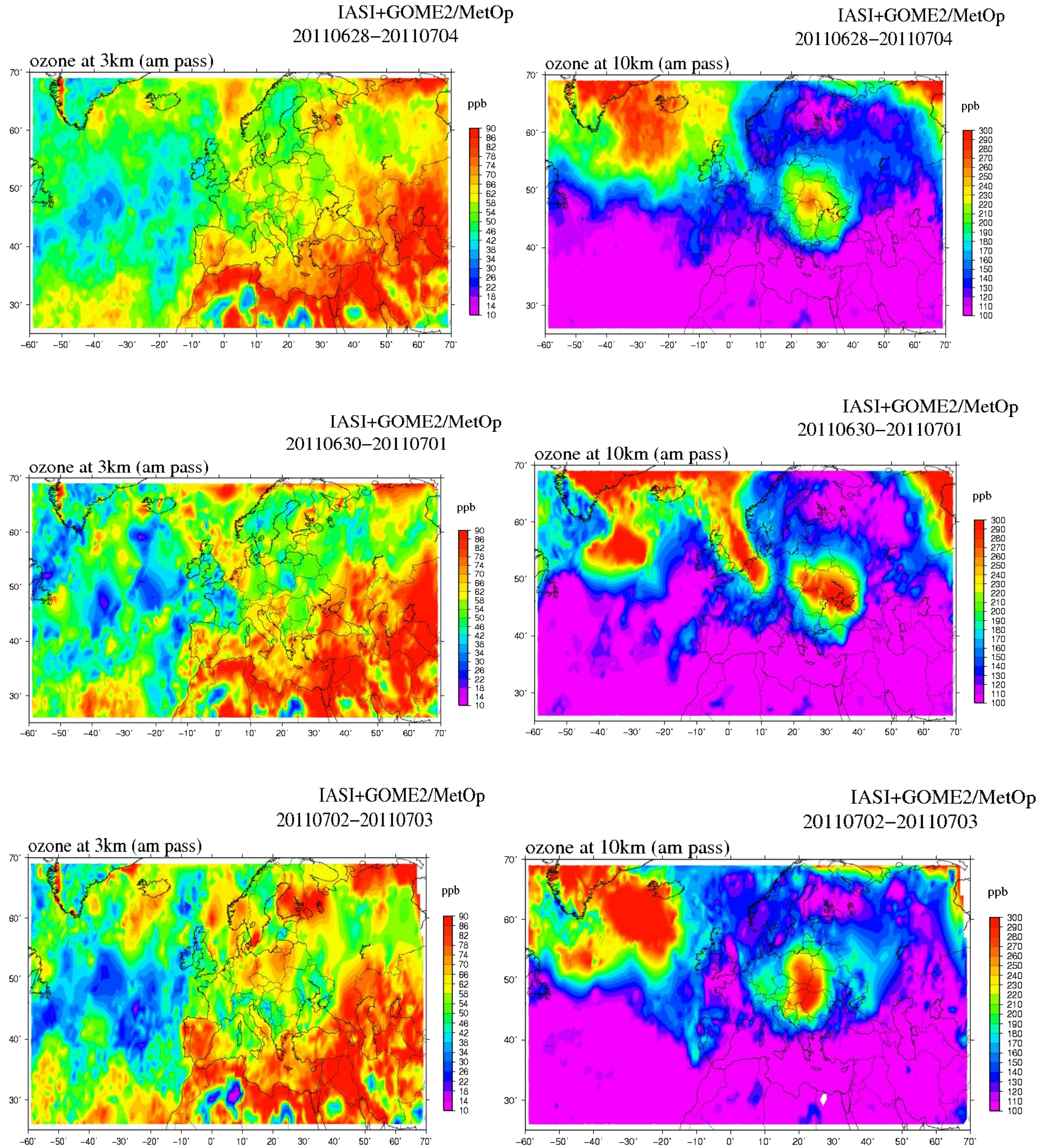
IASI+GOME2/MetOp
20110628−20110704
ozone at 3km (am pass)
ppb
IASI+GOME2/MetOp
20110628−20110704
ozone at 10km (am pass)
ppb
IASI+GOME2/MetOp
20110630−20110701
ozone at 3km (am pass)
ppb
IASI+GOME2/MetOp
20110630−20110701
ozone at 10km (am pass)
ppb
IASI+GOME2/MetOp
20110702−20110703
ozone at 3km (am pass)
ppb
IASI+GOME2/MetOp
20110702−20110703
ozone at 10km (am pass)
ppb

Fig. 11:

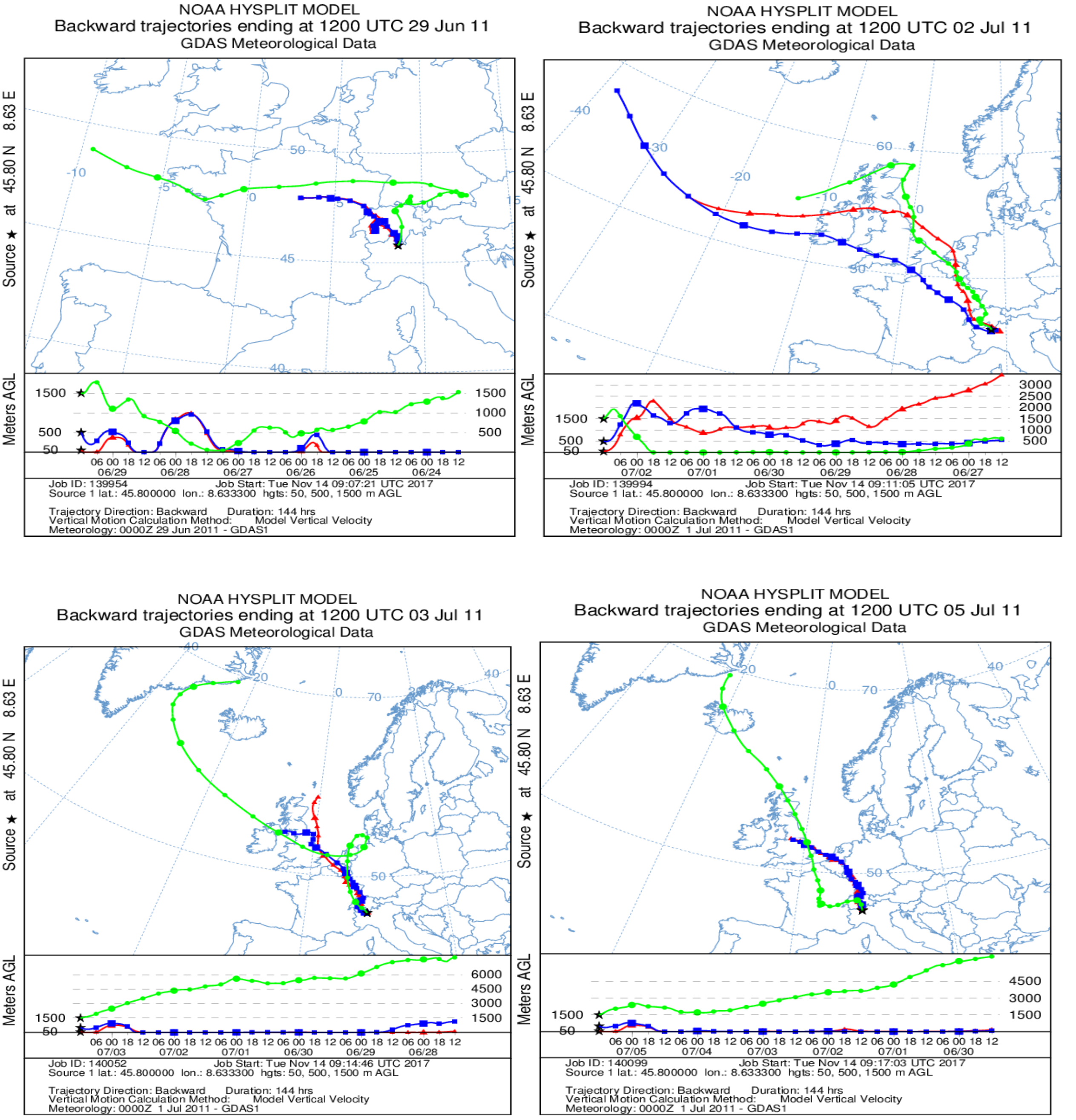
NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 29 Jun 11
GDAS Meteorological Data
Source ★ at 45.80 N 8.63 E
Meters AGL
Job ID: 139954 Job Start: Tue Nov 14 09:07:21 UTC 2017
Source 1 lat.: 45.800000 lon.: 8.633300 hgts: 50, 500, 1500 m AGL
Trajectory Direction: Backward Duration: 144 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 29 Jun 2011 - GDAS1
NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 02 Jul 11
GDAS Meteorological Data
Source ★ at 45.80 N 8.63 E
Meters AGL
Job ID: 139994 Job Start: Tue Nov 14 09:11:05 UTC 2017
Source 1 lat.: 45.800000 lon.: 8.633300 hgts: 50, 500, 1500 m AGL
Trajectory Direction: Backward Duration: 144 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 1 Jul 2011 - GDAS1
NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 03 Jul 11
GDAS Meteorological Data
Source ★ at 45.80 N 8.63 E
Meters AGL
Job ID: 140052 Job Start: Tue Nov 14 09:14:46 UTC 2017
Source 1 lat.: 45.800000 lon.: 8.633300 hgts: 50, 500, 1500 m AGL
Trajectory Direction: Backward Duration: 144 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 1 Jul 2011 - GDAS1
NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 05 Jul 11
GDAS Meteorological Data
Source ★ at 45.80 N 8.63 E
Meters AGL
Job ID: 140099 Job Start: Tue Nov 14 09:17:03 UTC 2017
Source 1 lat.: 45.800000 lon.: 8.633300 hgts: 50, 500, 1500 m AGL
Trajectory Direction: Backward Duration: 144 hrs
Vertical Motion Calculation Method: Model Vertical Velocity
Meteorology: 0000Z 1 Jul 2011 - GDAS1

Fig. 12:

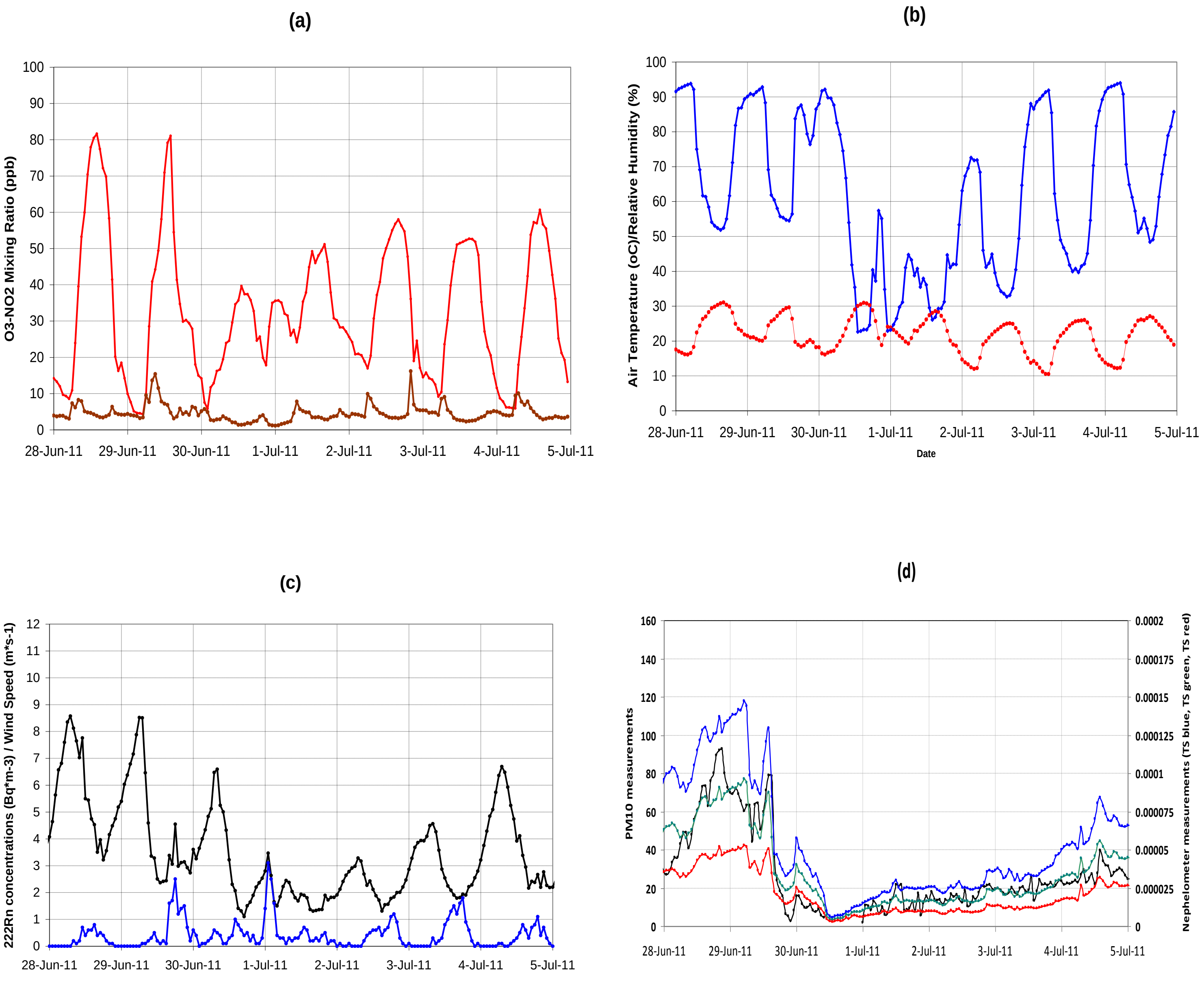
(a)
O3-NO2 Mixing Ratio (ppb)
100
90
80
70
60
50
40
30
20
10
0
28-Jun-11
29-Jun-11
30-Jun-11
1-Jul-11
2-Jul-11
3-Jul-11
4-Jul-11
5-Jul-11
(b)
Air Temperature (oC)/Relative Humidity (%)
100
90
80
70
60
50
40
30
20
10
0
28-Jun-11
29-Jun-11
30-Jun-11
1-Jul-11
2-Jul-11
3-Jul-11
4-Jul-11
5-Jul-11
Date
(c)
222Rn concentrations (Bq*m-3) / Wind Speed (m*s-1)
12
11
10
9
8
7
6
5
4
3
2
1
0
28-Jun-11
29-Jun-11
30-Jun-11
1-Jul-11
2-Jul-11
3-Jul-11
4-Jul-11
5-Jul-11
(d)
PM10 measurements
160
140
120
100
80
60
40
20
0
Nephelometer measurements (TS blue, TS green, TS red)
0.0002
0.000175
0.00015
0.000125
0.0001
0.000075
0.00005
0.000025
0
28-Jun-11
29-Jun-11
30-Jun-11
1-Jul-11
2-Jul-11
3-Jul-11
4-Jul-11
5-Jul-11

Fig. 13: