Reply to the reviewers' comments on the ACPD manuscript "Source attribution of European surface O3 using a tagged O3 mechanism" by Aurelia Lupaşcu and Tim Butler

Below we address the comments of the reviewers and questions raised during the open discussion of the manuscript "Source attribution of European surface O3 using a tagged O3 mechanism". We thank to the two Anonymous Reviewers for the time and effort reviewing the manuscript. We believe it has improved thanks to their comments. We have listed all reviewers' comments below and our answers are provided in blue. We also append a "track-changes" version of the revised manuscript as an appendix with all changes to the manuscript highlighted.

Response to Anonymous Referee #2 comments

This is a well-written paper that provides a unique analysis of all of the source contributions to surface ozone in Europe over the spring to autumn months in 2010. By creating additional tracers of NO, NO2 and their reservoirs to the chemical mechanism, the sources of ozone can be quantified without perturbing emissions (which changes the chemistry), as is frequently done for such analyses. This paper discusses in a comprehensive and clear manner the various contributions and their seasonal variation. A nice analysis of standard metrics (MDA8, W126, AOT40 and SOMO35) is also presented. I recommend publication after addressing the minor comments outlined below.

We thank the reviewer for their positive comments. Below you can find our point-by-point responses to all the issues raised.

1. There are numerous grammar errors (missing articles, plural/singular mismatch,etc.) that should be corrected.

Thank you for the useful feedback. As suggested by both reviewers, the manuscript has been carefully revised and the grammatical, linguistic and spelling mistakes in the manuscript have been carefully checked and corrected.

2. In Section 3.1 more should be said about how similar this simulation is to the one evaluated in Mar et al. (2016) to assure the reader that the results of that evaluation are really relevant here.

Following the reviewer's suggestion, we updated the sentence as follows "An extensive evaluation of WRF-Chem using the MOZART chemical mechanism to predict long term meteorological data and O3 levels has been presented previously (Mar et al., 2016). The main differences between the set-up up used in this study and the model described by Mar et

al. (2016) include differences between the versions of the model used (3.7.1 vs. 3.5.1, respectively), horizontal resolutions (50kmx50km vs. 45x45km, respectively), microphysics (Morrison vs. Lin, respectively) and cumulus schemes (Grell-Freitas vs. Grell 3-D, respectively), simulation years (2010 vs. 2007, respectively), anthropogenic emissions inventory (TNO-MACC III vs. TNO-MACC II, respectively), and chemical input and boundary conditions (extended CAM-Chem version 1.2 with MOZART-4 vs. MOZART-4/GEOS-5 simulations found at http://www.acom.ucar.edu/wrfchem/mozart.shtml, respectively)."

3. The table captions in the supplement should more clearly explain the contents (units,etc.)
Following the reviewer's suggestion, we updated the Tables caption as it follows: "Table 1. Percent contribution of local, European, long-range transported (LRT) and natural emissions sources to MDA8 O3 (ppb) at each receptor region during late spring, summer and early autumn 2010" and "Table 2. Percent contribution of different emission sources and types to total O3 at each receptor region as calculated for health and vegetation metrics. The metrics analysed are mean, MDA8 and 95th percentile (ppb) for the early "F" and late "S" simulation period, W126 (ppm-hours), SOMO35 and AOT40 (ppb-hours)".

Response to Anonymous Referee #3 comments

This is a very nice analysis that provides a lot of useful information and insight regarding the sources of ozone across Europe. The material is appropriate for ACP and I could recommend the paper for publication after it is revised according to my comments below.
We thank the reviewer for their constructive and valuable comments. Following the Reviewer's suggestion, we revised the paper and addressed all the concerns raised by providing response to individual comments below. Our responses are in blue.

General comments: 1) My only concern about the analysis is with regards to the ALPS region. This region combines the high elevations of the Alps (strong influence from long range transport of European and intercontinental origin) with the low elevations of the Po Valley, which is shielded from long-range transport by the Alps and experiences localized and intense air pollution episodes. Given the high variability of source regions and the high variability of local emissions, I don't think that any clear conclusions can be drawn for this region. The authors need to split this region into two parts: 1) the Po Valley; and 2) regions above 1500 m elevation. Then we should see that the high elevations have the greatest impact from long-range transport and the low elevations have the strongest impact from local emissions.

Following the recommendation of the reviewer, we have split the ALP source region into two receptor regions: the Po Valley and the high Alps. The manuscript was revised to account for these regions and the discussion was updated accordingly in the following sections:

- 2.2 (Experimental setup) by adding the following sentences “Except for ALP, the source regions within the European domain are identical to receptor regions. Given the complex topography of the ALP source region, we split this region into two receptor regions: the Po Valley region and the high Alps (regions above 1500 m elevation)”.
- 3.2 (Contribution of tagged precursor sources to the MDA8 O3 concentration) by recalculating the contribution of O3 coming from different sources and types to total O3 mixing ratio in the Po Valley and the high Alps receptor regions. We noticed that the contribution of different sources to the total MDA8 O3 mixing ratio in the Po Valley and the (old) ALP receptor regions give fairly similar results. Therefore, the text has been updated by using the newly calculated contributions in the Po Valley receptor region instead of ALP.
- 3.3 (Tagged ozone precursor contributions to exceedances of MDA8 target values – case study) by updating the manuscript as follows: “The relative contribution of emissions from different source regions to modelled MDA8 O3 and to observed MDA8 O3 values, after being scaled to account for the contribution of modelled sources of O3 types is generally similar for Po Valley and GEN receptor regions (see Fig. 6). In the Po Valley, we can pinpoint the main remote contributor as being MBS (see Fig. 6), followed by GEN, and FRA, suggesting a dominant westerly and northerly air flow. The recirculation of air masses in the Gulf of Genoa could accentuate the sea breeze and therefore more O3 coming from NOx associated with shipping activities in the Mediterranean will be transported to the coastal and inland station. The high Alps receptor region is less influenced by ALP emissions than the Po Valley, and is more influenced by remote sources (see Fig. 6). The increased contribution of O3 from CEN, ITA and FRA to both exceedance and non-exceedance days in the high Alps receptor region compared with the Po Valley receptor region highlights the impact of the transboundary transport of O3 and its precursors. Furthermore, the contribution of stratospheric as well as long-range sources was generally 6 % higher in this receptor region than in the Po Valley receptor region.”
- 3.4 (Tagged ozone precursor contributions to regulatory ozone metrics). We have recalculated the contribution of O3 coming from different sources and types to diverse health and vegetation metrics in the Po Valley and the high Alps receptor regions. Thus, in the revised manuscript, the comparison of different ozone metrics highlights the modelled values in the Po Valley instead of the ALP receptor region.

- We have updated the discussion in the Conclusion as follows: “To better understand the origin of MDA8 O3 exceedances, we compared modelled and observed values of MDA8 O3 concentration in the Po Valley, high Alps, Germany, and Benelux receptor regions. Throughout days exceeding the recommended thresholds of 120 µg/m^{-3}, the contribution from local sources was ~41 %, 34 % and 38 % of modelled MDA8 O3 for Po Valley, high Alps, and GEN, respectively. Throughout days not exceeding recommended thresholds, local emissions explain ~27 %, 16 % and 23% of modelled MDA8 O3 for the Po Valley, high Alps, and GEN, respectively. Moreover, this tagging approach revealed that the main remote sources of MDA8 O3 are MBS, GEN, and FRA for the Po Valley receptor region, and are FRA, CEN and UKI for Germany and Benelux receptor region. In addition, these analyses identified a persistently high contribution of transboundary sources to background O3 concentration in the high Alps receptor region”.

We have also updated the tables in the Supplementary material accordingly. The details of the changed text could be also seen in the tracked changes version of the manuscript, appended here.

2) In general the standard of English is fairly good but it needs a lot of polishing. There are too many instances of grammatical errors or awkward phrasing for me to list individually, but here are a few examples from the first paragraph of the Introduction: “The World Health Organization air quality guideline report that high O3 concentrations can cause damages to humans and vegetation” “Moreover, it has been shown that tropospheric O3 also affects radiative forcing and therefore contributing to climate change.” “To maintain a good air quality and understand O3’s response to climate change, it is important to understand the contribution of different sources of its precursors to the tropospheric O3 concentration.” Another example is this sentence in the Conclusions, which is difficult to understand: “Thus, we have seen that during the exceedances days, the contribution from local sources sources is~45 % and 38 % of modeled MDA8 O3, whilst during nonexceedances values is~32 % and 2 3% for ALP, respectively GEN.”

Thank you for your kind suggestion. The manuscript has been carefully revised and the grammatical, linguistic and spelling mistakes in the manuscript have been carefully checked and corrected. Following the reviewer’s suggestion, we have updated the introduction as follows “Ground-level O3 is an important air pollutant that damages human health (Fleming et al., 2018) and vegetation (Mills et al., 2018). It also affects the radiative forcing (e.g. Ramaswamy et al., 2001; Stevenson et al., 5 2013), and therefore contributes to climate change. Impacts of O3 on human health are associated with lung disease, chronic disease, and death from respiratory ailments. To protect human populations from exposure to high

levels of O3, the World Health Organization (WHO, 2006, 2017) recommended an air quality guideline for ozone in which the maximum daily average 8-h (MDA8) for O3 should not exceed 100 µg/m^{-3}. The European Environmental Agency (EEA, 2017a) reported that the EU long-term objective concentration of 120 µg/m^{-3} is often exceeded and that more than 90 % of the urban population of the European Union was exposed to O3 levels higher than the stricter recommendation set by the WHO". "To improve the air quality in certain areas, it is important to know the extent to which different precursors (NOx and VOCs) contribute to tropospheric O3 concentrations".

We realize that the sentence in the Conclusions creates confusion, therefore, we have rephrased this as follows: "To better understand the origin of MDA8 O3 exceedances, we compared modelled and observed values of MDA8 O3 concentration in the Po Valley, high Alps, Germany, and Benelux receptor regions. Throughout days exceeding the recommended thresholds of 120 µg/m^{-3}, the contribution from local sources was ~41 %, 34 % and 38 % of modelled MDA8 O3 for Po Valley, high Alps, and GEN, respectively. Throughout days not exceeding recommended thresholds, local emissions explain ~27 %, 16 % and 23% of modelled MDA8 O3 for the Po Valley, high Alps, and GEN, respectively."

Specific Comments:

Page 2, line 5 Here you state: "It has been shown that the background O3 concentrations have increased during the last several decades due to the increase of overall global anthropogenic emissions of O3 precursors (HTAP, 2010)" What is meant by background? HTAP uses the term global background to refer to natural ozone that would exist in the absence of anthropogenic emissions. This quantity cannot be measured but must be calculated by global models. Do you really mean to say that the global natural background has increased? Or do you mean that average observed global ozone has increased? According to the extensive review conducted by TOAR-Climate [Gaudel et al., 2018] the current in situ ozone monitoring network is insufficient to quantify ozone changes on the global or hemispheric scale; the available satellite products disagree on the trend, with some showing increases and some showing decreases.

We meant the surface ozone without contribution from local anthropogenic sources, which should more correctly be referred to as the "baseline" ozone. The text has been changed accordingly as follows: "A 2010 report from HTAP (HTAP, 2010) shows that the observed baseline O3 concentrations (concentrations without the contribution from local anthropogenic emissions) have increased throughout the last several decades since overall global anthropogenic emissions of O3 precursors have increased. However, a more recent study by Gaudel et al. (2018) has established that the global surface O3 trends exhibit high variability, and depend on several factors such as season, region, elevation and proximity to fresh

ozone precursor emissions. However, since the network capable of monitoring ozone levels is sparse, it is difficult to quantify the O3 changes on a global scale. Satellite-derived O3 measurements can be used to quantify changing levels of O3, but Gaudel et al. (2018) showed that these products are not capable of quantifying significant trends."

Line 6 Here you want to demonstrate that ozone air pollution is a current issue, but you only provide one reference that is now 11 years old and only applies to China. Please find additional references that are more current and cover other regions of the world. TOAR has recently published three papers that report present-day ozone using metrics relevant to human health [Fleming et al., 2018], vegetation [Mills et al., 2018] and climate [Gaudel et al., 2018]. These papers also provide up-to-date reviews of the literature concerning the impacts of ozone on humans, vegetation and climate. These papers would be helpful for the Introduction.

Following the reviewer's suggestion, we revised the sentence as it follows: "Surface O3 pollution due to urbanization and motorization processes are serious challenges for large cities (e.g. Chan and Yao, 2008; Folberth et al., 2015; Li et al., 2017, 2019). Paoletti et al. (2014) showed that in Europe and the United States of America, the average O3 concentration in the cities has increased at a faster rate than those observed in rural areas. Fleming et al. (2018) showed that the 4th highest daily maximum 8-hour O3 (4MDA8) are more ubiquitous at urban sites than at non-urban sites. This leads to a worsening of general air quality that, ultimately, affects human health and ecosystems (Paoletti et al., 2014; Monks et al., 2015; WHO, 2017; Fleming et al., 2018; Mills et al., 2018)."

In general the standard of English is fairly good but it needs some polishing, for example the following phrases in the first paragraph of the Introduction need more work: "The World Health Organization air quality guideline report that high O3 concentrations can cause damages to humans and vegetation" "Moreover, it has been shown that tropospheric O3 also affects radiative forcing and therefore contributing to climate change" To maintain a good air quality and understand O3's response to climate change, it is important to understand the contribution of different sources of its precursors to the tropospheric O3 concentration." Another example is this sentence in the Conclusions, which is difficult to understand: "Thus, we have seen that during the exceedances days, the contribution from local sources sources is ~45 % and 38 % of modeled MDA8 O3,whilst during nonexceedances values is~32 % and 2 3% for ALP, respectively GEN."

The reviewer has apparently made a duplicate comment since s/he had referred to these phrases in the "General comments" section, which we have already responded to.

Page 3, Line 21 Lefohn and Musselman stated that the W126 index ". . . would provide a more appropriate target for air quality management programs designed to reduce emissions from anthropogenic sources contributing to O3 formation". I'm not sure why this quote is provided. It seems to imply that W126 is a better metric then AOT40, but there is no agreement among the scientists who study the impacts of ozone on vegetation as to which metric is best. W126 was developed for a few limited crops and trees and is not necessarily applicable to other types of vegetation. For example, Harmens et al. [2018] found that a particular type of wheat is not sensitive to ozone peaks, which means W126 is not the best metric. Mills et al. [2018] give an overview of various vegetation metrics and they point out that the flux-based methods are the most accurate, but TOAR did not include these methods because their calculation on the global scale is not yet feasible.

Considering the reviewer's comment, we have simplified the paragraph for clarity. We have deleted the information less relevant to our manuscript since conflicts with other studies on this topic.

Page 4, Line 4 "Butler et al. (2018) describes in detail" should be "Butler et al. (2018) describe in detail" Because Butler et al. indicates many people, not one.

Corrected, thank you

Page 4, line 18 I don't understand this phrase: "has been modified to enable to model capacity to be used"

Following the reviewer's comment, we re-wrote the phrase as follows: "To overcome these limits, we modified the header file gdata.h, located in ~/KPP/kpp/kpp-2.1/src. Hence, the new gdata.h file considers a large number of species and reactions associated with this new chemistry option."

Page 4, Line 19 Should this sentence be a part of the preceding paragraph?

Indeed. Thanks to the reviewer for pointing to this.

Page 5 line 31 Here and throughout you need to be consistent with regards to the term, ozone concentration. "Ozone concentration" is appropriate when using units of μg m−3, but when using units of ppb, the correct term is "ozone mixing ratio"

We corrected, thank you.

In many places in the paper the term "NOx precursors" is used. This seems to be a redundant phase. Just define NOx as a precursor gas at the beginning of the paper and then afterwards just use "NOx" as it will be clear that it is a well-known precursor.

Following the reviewer's suggestion, in the "Introduction" we define NOx precursors as NOX and the use of the "NOx" term to refer to this precursor throughout the manuscript.

Page 8, line 18 "In all receptor regions, the MDA8 O3 concentration is dominated by O3 produced by remote anthropogenic precursors" I think this is overstated because it gives the impression that far more than half of the ozone is from remote anthropogenic precursors. But the range is 30-53%. Please re-phrase this sentence.
Following the reviewer's comment, we re-phrase it as follows "In all receptor regions, local anthropogenic sources have a lower contribution to MDA8 O3 mixing ratios than the sum of ozone due to anthropogenic sources in other European source regions and long-range transport of ozone from intercontinental source regions. The contribution of intercontinental transport to the total MDA8 O3 mixing ratio in Europe is consistent with previously reported results, i.e. Fiore et al. (2009) and Karamchandani et al. (2017), while this study allows us to identify which anthropogenic sources exert a strong influence on MDA8 O3 predicted in different regions.

Page 8 line 25 This sentence seems out of place and it should appear before the discussion of the stratospheric source.
Following the reviewer's suggestion, we moved this sentence.

Page 8 line 29 "Arabic Saudi peninsula" should be Arabian Peninsula
Corrected, thank you

Page 9 line 11 This statement is not quite correct: "Another consequence of enhanced photochemical activity during the summer season is the reduction of stratospheric O3" It should say: "Another consequence of enhanced photochemical activity during the summer season is that it reduces the relative influence of stratospheric O3" But in addition to changing the relative impact, do the absolute values of stratospheric ozone also diminish in summer, relative to spring?
Indeed, the absolute ozone mixing ratio attributable to the stratosphere is reduced in our simulations in summer compared with spring. We have updated the sentence as follows: "Another consequence of enhanced photochemical activity during summer is that it reduces the influence of stratospheric O3 from a domain-wide mean MDA8 O3 mixing ratio of 4.4 ppb in the spring to 1.3 ppb in the summer (Figs. 2 and 3)."

Page 9 line 31 Is OCN in these studies defined in the same way as in your study? If not then OCN should not be used when referencing the other studies.

No, in these studies the total shipping emissions are assigned to OCN. In our study, we define three source regions associated with the shipping activities: OCN (Oceanic sources coming from boundaries and the Atlantic Ocean), MBS (the Mediterranean and the Black Sea) and BNS (Baltic and the North Sea). Following the reviewer's suggestion, we remove these references.

Page 12, line 34 I don't understand what is meant by "efficiently complementing". I think you are trying to say that they behave similarly.

Thanks for this suggestion. We change accordingly the text "Since the difference between AOT40 and SOMO35 is only a few percentage points, regardless of the receptor region, we were able to conclude that they behave similarly, according to thresholds used to define these metrics."

Page 13, line 21 Here you say that W126 is more sensitive to local emissions because it does not include a threshold. But, in addition, isn't it also more sensitive to local emissions because this metric places much more weight on the high ozone values and high ozone is likely to be produced under hot stagnant conditions when local emissions are more important?

We agree that this sentence was misleading, so we have removed it from the manuscript. We would also like to point out that on pages 13, line 32 of the original manuscript we have a paragraph that provides an explanation to the observed sensitivity of the W126 metric to higher ozone concentrations. This paragraph states "this behaviour is closely linked to the definition of these metrics. If the O3 mixing ratio is less than 40 ppb, W126 has a weighting factor lower than 0.03, while AOT40 has a weighting factor of 0. Above this threshold, AOT40 has a weighting factor of 1, while in the case of W126 only O3 values higher than 100 ppb have a weighting factor of 1. Due to way these metrics are defined, predicted O3 values in each grid cell are accounted for the W126, may not be accounted for the AOT40 index".

Source attribution of European surface O_3 using a tagged O_3 mechanism

Aurelia Lupaşcu[1] and Tim Butler[1,2]

[1]Institute for Advanced Sustainability Studies (IASS), Potsdam, 14467, Germany
[2]Freie Universität Berlin, Institut für Meteorologie, Berlin, Germany

Correspondence: A.Lupascu (Aurelia.Lupascu@iass-potsdam.de)

Abstract.

Tropospheric ozone (O_3) is an important air pollutant that affects human health, ecosystems, and climate. The contributions of O_3 precursor emissions from different geographical source regions to the O_3 concentration can help to quantify the effects of local versus remote transported precursors on the O_3 concentration in a certain area. This study presents a "tagging" approach within the WRF-Chem model that attributes O_3 concentration in several European receptor regions to nitrogen oxides (NO_x) emissions from within and outside of Europe during April-September 2010. We also examine the contribution of these different precursor sources to various O_3 metrics ~~,~~and their exceedance events. Firstly, we show that the spatial distributions of simulated monthly mean MDA8 from tagged O_3 sources regions and types for late spring, summer and early autumn 2010 varies with season. For summer conditions, O_3 production is dominated by national and intra-European sources, while in the late spring and early autumn intercontinental transported O_3 is an important contributor to the total O_3 concentration. We have also identified shipping activities in the Mediterranean Sea as an important source of O_3 for the Mediterranean countries, as well as the main contributor to high modelled MDA8 O_3 concentration ~~, modelled~~ in the Mediterranean ~~basin~~ Basin itself. Secondly, to have a better understanding of the origin of MDA8 O_3 exceedances, we compare modelled and observed values of MDA8 O_3 concentration in the "~~Alps~~Po Valley" and "Germany-Benelux" receptor regions, revealing that the contribution from local sources is about ~~45~~ 41 % and 38 % of ~~modeled~~ modelled MDA8 O_3 during the exceedances days respectively. By examining the relative contributions of remote NO_x sources to modelled and observed O_3 exceedance events, we determine that model underrepresentation of long-range O_3 transport could be contributing to a general underestimation of modelled O_3 exceedance events in the Germany-Benelux receptor region. Thirdly, we quantify the impact of local vs. ~~non local~~ non-local NO_x precursors on O_3 production for each European receptor region using different O_3 metrics. The comparison between mean, MDA8 and ~~95th~~ 95^{th} percentile O_3 metrics accentuate the importance of large contributions from locally-emitted NO_x precursors to the high-end of the O_3 distribution. When we compare the vegetation and health metrics, we notice that the SOMO35 and AOT40 indexes exhibit ~~a~~ rather similar behaviour, while the W126 index accentuates the importance of local emissions. Overall, this study highlights the importance of a tagging approach to quantify the contribution of local and remote sources to the MDA8 O_3 concentration during several periods as well to different O_3 metrics. Moreover, this method could be applied to assess different mitigation options.

1 Introduction

Tropospheric ozone (O_3) is formed primarily through reactions ~~of~~ between nitrogen oxides (NO_x) and volatile organic compounds (VOC) that occur in the presence of sunlight. ~~The~~ Ground-level O_3 is an important air pollutant that damages human health (Fleming et al., 2018) and vegetation (Mills et al., 2018). It also affects the radiative forcing (e.g. Ramaswamy et al., 2001; Stevenson , and therefore contributes to climate change. Impacts of O_3 on human health are associated with lung disease, chronic disease and death from respiratory ailments. To protect human populations from exposure to high levels of O_3, the World Health Organization (WHO, 2006, 2017) recommended an air quality guideline ~~(WHO, 2006, 2017) report that high~~ for ozone in which the maximum daily average 8-h (MDA8) for O_3 ~~concentrations can cause damages to humans and vegetation. It has been shown that the background~~ should not exceed 100 μg m^{-3}. The European Environmental Agency (EEA, 2017a) reported that the EU long-term objective target concentration of 120 μ is often exceeded and that more than 90 % of the urban population of the European Union was exposed to O_3 levels higher than the stricter recommendation set by the WHO. A 2010 report from HTAP (HTAP, 2010) shows that the observed baseline O_3 concentrations ~~have increased during~~ (concentrations without the contribution from local anthropogenic emissions) have increased throughout the last several decades ~~due to the increase of~~ since overall global anthropogenic emissions of O_3 precursors ~~(HTAP, 2010). Large cities are facing serious challenges in~~ have increased. However, a more recent study by Gaudel et al. (2018) has established that the global surface O_3 trends exhibit high variability, and depend on several factors such as season, region, elevation and proximity to fresh ozone precursor emissions. However, since the network capable of monitoring ozone levels is sparse, it is difficult to quantify the O_3 changes on a global scale. Satellite-derived O_3 measurements can be used to quantify changing levels of O_3, but Gaudel et al. (2018) showed that these products are not capable of quantifying significant trends. Surface O_3 pollution due to urbanization and motorization processes ~~(e.g. Chan and Yao, 2008). Moreover, it has been shown that tropospheric~~ are serious challenges for large cities (e.g. Chan and Yao, 2008; Folberth et al., 2015; Li et al., 2017, 2019). Paoletti et al. (2014) showed that in Europe and the United States of America, the average O_3 concentration in the cities has increased at a faster rate than those observed in rural areas. Fleming et al. (2018) showed that the 4th highest daily maximum 8-hour O_3 ~~also affects radiative forcing (e.g. Ramaswamy et al., 2001; Stevenson et al., 2013) and therefore contributing to climate change. To maintain a good air quality and understand O_3's response to climate change~~(4MDA8) are more ubiquitous at urban sites than at non-urban sites. This leads to a worsening of general air quality that, ultimately, affects human health and ecosystems (Paoletti et al., 2014; Monks et al., 2015; WHO, 2017; Fleming et al., 2018; Mills et al., 2018). To improve the air quality in certain areas, it is important to ~~understand the contribution of different sources of its~~ know the extent to which different precursors (NO_x and ~~VOC) to the~~ VOCs) contribute to tropospheric O_3 ~~concentration~~concentrations.

~~Emissions~~ Information regarding levels of NO_x and ~~VOCs, as well as the emissions sources~~ VOC emissions and weather conditions ~~are important to understand~~ enhance our ability to predict the formation of ~~the~~ tropospheric O_3 ~~in the troposphere. The~~ . The continuous development of chemical transport models ~~leads~~ can lead to a better understanding of the processes that contribute to ~~high O~~high-O_3 episodes~~and can help the authorities to develop strategies to reduce the impact of O_3 on both human well-being and ecosystems by knowing the impact of source emissions of~~ . Knowing the impacts of NO_x and VOC

emissions from sources such as surface anthropogenic ~~sources~~activities, fires, soil, and the stratosphere on total O_3 production can help authorities develop strategies aimed at reducing the impact of high levels of O_3 on the well-being of both humans and ecosystems. Several approaches have been used to determine the ~~source attribution~~ extent to which individual sources contribute to total levels of O_3. For example, ~~estimations of the changes in O_3 concentration have been made by~~ perturbation of different ~~emissions categories~~ emission categories have allowed scientists to make an estimations regarding the contributions of individual sources of O_3 to total O_3 levels (e.g. Fiore et al., 2009).

Tagging techniques have also been used in modelling studies to determine source~~attributions for pollutants~~ /receptor relationships and how individual sources of pollutants contribute to total pollution levels at given locations~~and source/receptor relationships~~. Pollutants with relatively low chemical reactivities, such as carbon monoxide (CO), can be "tagged" according to ~~its~~ their emission sectors or regions for attribution studies (e.g. Pfister et al., 2011). Sudo and Akimoto (2007), and Derwent et al. (2015) used O_3 tracers tagged by their region of formation ~~. They have found that the~~ to show that intercontinental transport of O_3 can occurring from polluted source ~~region~~ regions, such as North America and East-Asia, appears to be the most important source of tropospheric O_3 in Europe. ~~The~~ Other studies, including those of Wang et al. (2009) and Grewe et al. (2010, 2012, 2017) ~~studies showed that the tagging method is used to identify contribution from individual sources due to its ability to track the~~ have used tagging methods to identify the contribution of individual sources of O_3 to overall levels. This method is especially useful since it can track emitted NO_x species during transport and chemical processing. Moreover, Grewe et al. (2012) showed the impact of the tagging method on mitigation measures, while Dahlmann et al. (2011) ~~studied~~ examined the contribution of O_3 sources to O_3 radiative forcing. ~~The studies of~~ Work by Emmons et al. (2012) and Butler et al. (2018) describe a procedure for tagging O_3 produced from NO_x sources through updates to the MOZART chemical mechanism~~. In addition, Butler et al. (2018) have extended~~ , and Butler et al. (2018) expanded the tagging technique to account for ~~the~~ VOC sources.

Based on ~~Emmons et al. (2012) work~~ the work of Emmons et al. (2012), Pfister et al. (2013) and Safieddine et al. (2014) ~~used~~ were able to use the WRF-Chem regional model to quantify the ~~role~~ contribution of inflow (~~defined as source of~~ tagged O_3 and odd nitrogen species entering into ~~a~~ the regional domain at the lateral boundaries) and of anthropogenic NO_x precursors (named NO_x in the following) on the surface O_3 levels. Using a slightly different methodology, Gao et al. (2016) have ~~employed~~ implemented within WRF-Chem ~~a tagging~~ framework a tagging method based on Ozone Source Apportionment Technology (OSAT) (Yarwood et al., 1996) incorporated in the Comprehensive Air quality Model with extensions (CAMx).

~~Many efforts have been made to understand~~ Much effort has been focused understanding the origin of tropospheric O_3 and the key role played by the intercontinental transport, the contribution of stratospheric O_3 intrusion, and of different emissions sources to tropospheric O_3 concentration in a wide range of receptor regions. ~~For a better understanding of these interactions,~~ To better understand how these reaction interact it is necessary to know the ~~relation between the amount~~ relationship between levels of an emitted species and ~~its~~ atmospheric concentration. ~~Thus we can~~ When this information is known, it is possible to quantify the contribution of different emission precursor sources to ~~the~~ overall O_3 concentration ~~at a~~ levels at a particular receptor location. For this purpose, ~~following~~ we followed a strategy outlined in Emmons et al. (2012) and Butler et al. (2018) ~~, we implemented~~ to implement a tagging technique into the regional WRF-Chem model~~a tagging technique that~~ . The model can

be used to quantify ~~the~~ source contributions to the tropospheric O_3 concentration, by "tagging" ~~emissions of~~ NO_x emissions, and corresponding ~~resulting products and following them to the~~ products so that they can be traced to the final production of O_3.

~~Important objective when we are studying~~ When studying the effects of O_3 ~~is its effect~~, the impact of the compound on humans and vegetation is of the utmost importance. Therefore, ~~based on hourly averaged data,~~ several exposure indexes have been defined ~~in order~~ to describe the relationship between O_3 and both human health and agricultural crop yield that are based on hourly averaged data. Musselman et al. (2006), Agathokleous et al. (2018), and Lefohn et al. (2018) ~~present a literature overview on~~ review literature describing O_3 metrics~~, while Paoletti et al. (2007) presents~~. Additionally, a work by Paoletti et al. (2007) has provided a list of common O_3 exposure metrics used to assess risk to human health and vegetation~~across Italy during the 2000-2004 period~~. Here we use some well-known O_3 metrics, such as MDA8, SOMO35, AOT40, and W126. The MDA8 index ~~(Lefohn et al., 2018) is~~ has been defined as the maximum daily average 8-h (MDA8) O_3 values (~~in units of~~ ppb) (Lefohn et al., 2018). SOMO35 (WHO, 2001) ~~is~~ has been determined by European protocols (EU directive 2008/50/EC, 2008) and ~~it~~ is defined as the annual sum of MDA8 O_3 with a cut-off of values of 35 ppb~~,~~. Both MDA8 and SOMO34 are health-related metrics. The AOT40 and W126 vegetation metrics have been used ~~in air pollution regulation in~~ to regulate air pollution in both Europe (EU directive 2008/50/EC, 2008) and the United States (U.S. EPA regulations https://www.gpo.gov/fdsys/pkg/FR-2015-10-26/pdf/2015-26594.pdf). ~~In the~~ European legislation (EU directive 2008/50/EC, 2008) ~~, the~~ AOT40 metric is ~~accumulated over the daytime period~~ measured throughout daytime periods from May to July ~~and it~~ (growth season) and has a defined target limit of 18000 μg m^{-3} h (9000 ppb $-$ hours) and a long term objectives of 6000 μg m^{-3} h (3000 ppb $-$ hours). A standard of 15 ppm $-$ hours ~~is~~ has been defined for the seasonal W126 index, which is averaged over three years. ~~Lefohn and Musselman (2012, https://www.fcpotawatomi.com/wp-content/uploads/2015/01/Vegetation.pdf) stated that the W126 index "… would provide a more appropriate target for air quality management programs designed to reduce emissions from anthropogenic sources contributing to O_3 formation".~~ These metrics have been used ~~side by side~~ to assess the impact of mitigation strategies (Avnery et al., 2013), the impact of industry on air quality management issues (Vijayaraghavan et al., 2016), and the impact of high O_3 levels and temperatures on crops (Tai and Val Martin, 2017).

In this paper, we use a tagged O_3 mechanism in the WRF-Chem ~~regional chemistry-climate~~ model to understand the ~~contributions of~~ contribution of emitted O_3 ~~precursor emissions~~ precursors from different geographical source regions and types on the modelled O_3 concentration in several European receptor regions. In Section 2 we briefly describe the details of ~~the implementation of~~ implementing this tagging technique~~pointing to the~~, and describe changes made to both the chemical mechanism and WRF-Chem code. Section 2 also ~~provides a description of~~ describes the WRF-Chem configuration, simulation design, and input data used in the study. An analysis of the WRF-Chem ~~simulations~~ simulation is presented in Section 3, while Section 4 summarizes our findings.

2 Model simulation

2.1 Tagging technique

~~In order to implement a~~ To perform a WRF-Chem model simulation using a tagging approach, several changes must be implemented in the model code to accommodate additional tracers and reactions representing tagged constituents. Butler et al. (2018) describes in detail how the tagging technique was implemented in the Community Earth System Model. The tagging technique used in this study is based on the same approach ~~,~~ and uses the same modified version of the MOZART chemical mechanism. Further detail on how the chemical mechanism was extended can be found in Butler et al. (2018).

~~In order to~~ To use the NO_x tagging mechanism, a new chemistry option was added in the namelist.input file: chem_opt=113 ~~as~~ and through the code. The coupling of the new chemical scheme with microphysics and radiative processes requires several modifications to the code: 1) The first step is to create a new chemistry option. The package mozart_tag_kpp (chemopt==113) has been added to ~~~~/WRFV3/Registry/registry.chem together with new model variables for tagged NO_x species ~~, for example,~~ (e.g. O3_X_INI, O3_X_STR, etc). For this purpose, the pre-processing software described in Butler et al. (2018) was adapted ~~in order~~ to produce a new chemical mechanism; 2) The new chemistry package is a KPP option. Therefore, we created a new subdirectory in ~~~~/WRFV3/chem/KPP/mechanisms/ directory containing the files (*.spc, *.eqn, *.kpp, and *.def) which defined the chemical model species and constants, chemical reactions in KPP format, model description, computer language, precision, and integrator.

The KPP chemical preprocessor, version 2.1 (Sandu and Sander, 2006) used by WRF-Chem ~~has an upper limit of~~ limits the numbers of species and reactions in the chemical mechanism. ~~Thus,~~ To overcome these limits, we modified the header file gdata.h, located in ~/~~chem/~~KPP/kpp/kpp-2.1/src~~, has been modified to enable to model capacity to be used for~~ . Hence, the new gdata.h file considers a large number of species and reactions ~~.~~

associated with this new chemistry option. Further, we updated the subroutines in the ~~chem directory to take in account these packages~~~/WRFV3/chem directory consider the new chemistry package. The modules that we modified are described in the Appendix.

Although WRF-Chem uses the Advanced Research WRF (ARW) dynamic core in this simulation which conserves mass and scalar mass (Grell et al., 2005), the tagged O_3 species are advected independently. Thus, numerical errors associated with the advection scheme led to gradients in the sum of tagged species concentration compared to the "real" concentration; therefore, the relationship between these variables is not conserved. Since the advection scheme fails to reproduce the expected solution (in which the sum of the tagged species concentration at each grid point must be equal to "real" concentration), we solve this by fixing all undershoots and/or overshoots assuming that the sum of tagged species mass is proportional to the "real" concentration. This technique was also applied in Flemming et al. (2015), and Gromov et al. (2010).

Compared to Pfister et al. (2013) and Safieddine et al. (2014) work, the expanded tagging technique used in this study has the advantage that multiple tags can be defined in each model run.

2.2 ~~Experiment~~ Experimental setup

WRF-Chem version 3.7.1 was used for this study to account for the impact of different global and European O_3 precursor source regions to several European receptor regions during the April-September 2010 period. A single domain, that covers the area between 32° N and 70° N, and 29° W and 57° E, was used with 50-km grid spacing and 35 vertically-stretched layers from the ground up to 50 hPa. The physics options used for this study include the Morrison double-moment microphysics scheme (Morrison et al., 2009), the Grell-Freitas cumulus parameterization (Grell and Freitas, 2014), the Rapid Radiative Transfer Model (Iacono et al., 2008) for longwave and Goddard shortwave scheme (Chou and Suarez, 1994), the Yonsei University boundary-layer parameterization (Hong et al., 2006), and the Monin-Obukhov scheme for the surface layer (Jiménez et al., 2012). The initial and boundary conditions for meteorological fields are ~~using~~ taken from the European Centre for Medium-Range Weather Forecasts (ECMWF) ~~reanalyses~~reanalysis. Anthropogenic emissions were obtained from the TNO-MACC III emission inventory for Europe (Kuenen et al., 2014). Because the model domain extends beyond the edges of the TNO-MACC III inventory, we used for completion emissions from the HTAP V2 inventory (http://edgar.jrc.ec.europa.eu/htap_v2). Biogenic emissions were computed on-line using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) model (Guenther et al., 2006). The biomass burning emissions are based on Fire INventory from NCAR (FINN) (Wiedinmyer et al., 2011).

For this WRF-Chem simulation, the tagged MOZART chemical mechanism for NO_x emissions (Butler et al., 2018) is used to represent the gas-phase chemistry. The photolysis rates were computed using the Fast Tropospheric Ultraviolet and Visible (FTUV) Radiation Model ~~(Tie et al., 2003; Li et al., 2005)~~(Tie et al., 2003; Li et al., 2005). The dry deposition was calculated following the Wesely (1989) resistance method, while the wet removal scheme for the tagged MOZART chemistry is based on Neu and Prather (2012).

~~In order to~~ To represent the impact of transported O_3 from different regions outside of the domain, ~~for the trace gases we used~~ we used chemical boundary conditions derived from the extended CAM-Chem version 1.2 global simulations. Butler et. al (in preparation) used the tagging approach within the CAM-Chem model for several HTAP2 source regions such as: ASI (Asia), NAF (North-Africa), NAM (North-America), OCN (Oceanic sources), RBU (Russia, Belarus, Ukraine), and RST (rest of the world), as well as for several source types: BIO (biogenic emissions), BMB (biomass burning emissions), LGT (lightning), and STR (stratospheric O_3). Using a division of source/receptor regions within the European model domain, we define 15 geographical regions for this study, as shown in Figure 1 and Table 1. ~~Source regions within the European domain are identical to European receptor regions in our study.~~ A similar division of European regions has been used by Christensen and Christensen (2007) and Otero et al. (2018) to address the main sources of uncertainty in regional climate simulations, as well as during the AQMEII project (i.e. Struzewska et al., 2015). Except for ALP, the source regions within the European domain are identical to receptor regions. Given the complex topography of the ALP source region, we split this region into two receptor regions: the Po Valley region and the high Alps (regions above 1500 m elevation).

For each receptor region, we analyse the impact of the anthropogenic NO_x emissions coming from different source regions to the total O_3 concentration. The BIO, BMB, LGT, and STR types are also included in the simulation, but without including them into the division of source/receptor regions.

2.3 Ozone metrics

~~By the means of different~~ Using different metrics to assess the impact of O_3 ~~impact metrics we determinate which are the most important O_3 precursor sources for different kinds~~, we can determine which precursor sources most highly influence the accumulation of O_3 ~~impact~~ in different receptor regions, and thus to provide ~~insight into appropriate mitigation measures~~ insights into which type of mitigation measures will be useful for a particular geographic area. These metrics include the mean O_3 concentration, the mean of ~~the maximum daily 8-hour O_3 (~~MDA8~~)~~, the cumulative exposure to mixing ratios above 35 ppb (SOMO35) (Colette et al., 2012), and the ~~95th~~ 95^{th} percentile for surface O_3~~, the impacts~~. Neither the impact of O_3 exposure on trees, plants and ecosystems (W126) (Lapina et al., 2014), nor the AOT40 accumulation metric (the threshold is 40 ppb) were used to assess risk to vegetation from O_3 exposure (UNECE, 2010).

The European Air Quality Directive (EU directive 2008/50/EC, 2008) ~~defines a target~~ specifies that O_3 exposure should remain below a target MDA8 O_3 value of 120 μg m^{-3} ~~for the MDA8 concentration~~, which can be exceeded up to 25 days per calendar year averaged over three years. The ~~modeled~~ modelled daytime AOT40 (during local daylight hours 8 AM – 7 PM) was calculated according to Equation (1).

$$AOT40 = \sum_{i=1}^{90\ \text{days}} \left(\sum_{h=8}^{19} max\left(\mathrm{O}_{3i,h} - 40{,}0\right) \right) \tag{1}$$

According to ~~the~~ European legislation (EU directive 2008/50/EC, 2008), the AOT40 metric is accumulated over the daytime period from May to July (growth season) and it has a defined target limit of 18000 μg m$^{-}3$ h (9000 ppb $-$ hours) and a long term objective of 6000 μg m^{-3} h (3000 ppb $-$ hours). W126~~is calculated~~, however, is described according to U.S. EPA regulations (https://www.gpo.gov/fdsys/pkg/FR-2015-10-26/pdf/2015-26594.pdf). A standard of 15 ppm $-$ hours is defined for the seasonal W126 index~~averaged over three years~~, which is an average over a three-year period. For this study, the hourly surface O_3 tagged outputs for April through September are used to calculate the highest 3-month W126 index values (see Eq. 2):

$$W126 = \sum_{i=1}^{90\ \text{days}} \left(\sum_{h=8}^{19} \mathrm{O}_{3i,h} \cdot \left(\frac{1}{1 + (4403 \cdot \mathrm{e}^{-126 \cdot \mathrm{O}_{3i,h}})} \right) \right) \tag{2}$$

According to Lefohn et al. (1988), the W126 index includes all hourly O_3 values within the specified time range, although a lower weight is given to hourly O_3 concentrations ~~bellow~~ below the inflection point ~~at~~ of 65 ppb, ~~whilst~~ while values above 90 ppb ~~the weighting factor is almost 1.~~ are weighted with a factor of almost one. SOMO35 (WHO, 2001) is defined as the sum of the MDA8 O_3 with a cut-off of 35 ppb (see Eq. 3). For this metric, the EU air quality directives do not prescribe a limit

or a target values.

$$SOMO35 = \sum_{h=1}^{6\ \text{months}} max\,(\text{MDA8}_i - 35{,}0.0) \tag{3}$$

The contribution of tagged O_3 ~~concentration to each metric~~ is based on formulations of each metric and is calculated from the model output~~based on metric formulations~~. In the case of the MDA8 and ~~95th~~ 95^{th} percentile metrics, we ~~search~~ searched for the specific period ~~when the calculated values for~~ in which calculated values of total O_3 concentration meet the requirements ~~given by~~ for the formulation of these metrics. Once this is identified, ~~the~~ tagged O_3 concentrations are extracted for the same period which ~~are then used further in our~~ can then be used for further analysis. However, the contribution of ~~these~~ concentration of tagged O_3 ~~concentrations to the cumulative metrics is slightly differentand it uses~~ on cumulative metrics are slightly different, a large proportion of each tagged species ~~to the~~ is used to determine total O_3, as illustrated below for AOT40 at ~~specific hour~~a specific time period:

$$AOT40_{tag} = \sum_{i=1}^{90\ \text{days}} max\left((\text{O}_3 - 40.) \cdot \frac{\text{O}_{3,tag}}{\text{O}_3}, 0.0\right) \tag{4}$$

Based on their formulation, we grouped ~~these metrics in~~ metrics into either non-cumulative (mean O_3, MDA8, and the ~~95th percentile) and~~ 95^{th} percentile) or cumulative (SOMO35, W126, and AOT40) categories. Since ~~these~~ the latter metrics have different ~~formulation (using~~ formulations (including hourly O_3 values ~~of O_3 values above a~~ above a specific threshold) and ~~they~~ do not cover the same ~~period of time~~periods, to facilitate a more direct comparison between findings from multiple O_3 metrics, an analysis of the relative contribution of different source regions to ~~the~~ total O_3 in each receptor region ~~using different O_3 metrics is~~ was determined. This was done using averaged values for non-cumulative metrics, ~~6-months~~ and 6-month sums for SOMO35, AOT40~~calculated for~~ , for cumulative metrics useful for evaluating effects on crops (cumulated over May-July period) and ~~maximum of 3-months~~ a maximum of 3-month sums for every consecutive ~~3-months period for~~ 3-month period determined using the W126 index.

3 Results and discussions

~~The~~ Our discussion of the ~~model results focuses~~ results of the model is focused on the April-September 2010 period. We first briefly evaluate the ability of WRF-Chem to reproduce ~~the~~ meteorological parameters using measurements from the Global Weather Observation (GWO) dataset provided by the British Atmospheric Data Center (BADC), and observed O_3 ~~concentration~~ concentrations using the measurements included in ~~the AirBase~~AirBase, a European air quality ~~data base (?)~~ database (EEA, 2017b). We then ~~provide~~ provided a more detailed analysis of the contribution of ~~the~~ different source regions and types to ~~the~~ MDA8 values describing total O_3 for the analysed period.

3.1 Evaluation of meteorology and chemistry

Since the accurate simulation of meteorological parameters represents a key factor ~~that affects the trace gases concentration~~affecting the concentrations of trace gases, we briefly compare the modelled mean sea level pressure (MSLP), 2 m temperature (T2M),

10 m wind speed (WS10M) and direction (WD10M) variables against ~~the~~ GWO measurement. Predicted model variables ~~are~~ were then evaluated against observations using statistical scores that include normalized mean bias (NMB), and the correlation factor between simulated and measured values (r).

An extensive evaluation ~~and discussion of the~~ of WRF-Chem using the MOZART chemical mechanism to predict long term meteorological data and O_3 ~~model performance of WRF-Chem using the MOZART chemical mechanism~~ levels has been presented ~~by Mar et al. (2016).~~ previously (Mar et al., 2016). The main differences between the set-up up used in this study and the model described by Mar et al. (2016) include differences between the versions of the model used (3.7.1 vs. 3.5.1, respectively), horizontal resolutions (50kmx50km vs. 45x45km, respectively), microphysics (Morrison vs. Lin, respectively) and cumulus schemes (Grell-Freitas vs. Grell 3-D, respectively), simulation years (2010 vs. 2007, respectively), anthropogenic emissions inventory (TNO-MACC III vs. TNO-MACC II, respectively), and chemical input and boundary conditions (extended CAM-Chem version 1.2 with MOZART-4 vs. MOZART-4/GEOS-5 simulations found at http://www.acom.ucar.edu/wrf-chem/mozart.shtm respectively).

Due to the coarse resolution of ~~the domainthat will not properly reproduce~~ our domain, the air parcel dynamics associated with the complex topography ~~in the mountainous areas , we assess~~ of mountainous areas was not properly reproduced. Thus, we assessed the ability of the model to reproduce the meteorological variables using only those ~~measurements sites located bellow~~ sites located below 1500 m above ~~ground~~ sea level. MSLP ~~is well reproduced at European scale~~ data was well reproduced over the entire period (NMB ~~of =~~ 0 % and r ~~of =~~ 0.98). ~~In terms of spatial correlation,~~ The model predicted T2M ~~performs very~~ values well (r = 0.91), however ~~the observed~~ temperature is underestimated by 3 % (see Table 2). WS10M ~~is~~ was also fairly well reproduced both in terms of spatial and temporal variability (NMB = 8 %, r = 0.63). Yet, WD10M ~~did not perform~~ data could not be predicted as well as ~~the~~ other meteorological variables (NMB = 13 %, r = 0.47)~~and this behavior~~ , behaviour could be related to the existence of unresolved topography features (Jimenez and Dudhia, 2012). ~~Also~~However, the model performance is similar to Mar et al. (2016) and Tuccella et al. (2012).

We also ~~compare the~~ compared modelled MDA8 O_3 ~~concentration against~~ concentrations with observations provided by the ~~publicly available~~ publicly-available AirBase dataset. The relatively coarse resolution of the domain ~~might~~ may not be representative ~~for the changes in the~~ of changes in local emissions when the measurements are ~~representative for urban condition; thereforefurther for the analysiswe use~~ taken from urban areas; therefore, to aid in the analysis, we used only those stations characterized as rural~~background~~. As can be seen ~~from~~ in Table 3, ~~the model evaluation over the whole period shows that both tests perform quite well in terms of concentration (NMB = -5.2 %)~~ evaluation of the model over entire period revealed that the model performs quite well with respect to the prediction of concentration and temporal evolution~~(r = 0.69)~~. Mar et al. (2016) reported a mean bias (MB) value of 15.85 μg m^{-3} and an NMB of 17% for the ~~June-August~~ June–August 2007 period when the MOZART mechanism was used to assess the chemical performances of the model~~, whereas for the same period we obtain a MB~~ . For the same time period, we obtained an MB value of -5.92 μg m^{-3} and ~~a NMB~~ an NMB value of -6.3%. Tuccella et al. (2012) reported an annual MB of -1.4 μg m^{-3} when the RADM2 chemical mechanism was used to simulate ~~the year~~ a period throughout 2007. Month-to-month analysis (Table 3) shows that the model reproduces the O_3 concentration well compared to the Mar et al. (2016) and Tuccella et al. (2012)~~studies~~. Even though the ~~model performance~~ performance of the model in

terms of temporal variation is ~~relavitely~~ relatively good (r values fall between 0.58 and 0.71), it mostly ~~underestimates the observed concentration~~underestimated concentrations of O_3, except in September, when the model ~~overestimates the observed~~ overestimated concentrations (NMB = 4.6%). ~~The modelled errors could~~ Errors of the model may be explained by a wide range of uncertainties related to ~~the~~ modelled physical and chemical processes such as grid resolution, vertical and horizontal transport, boundary layer mixing, ~~emissions~~ emission inventory, chemistry and photolysis rates, dry deposition~~and~~ , wet scavenging, etc~~as well as uncertainties in the measurements . As~~ . It is also possible that uncertainties in measurements contribute to observed errors. Since the focus of this study is on the contribution of different O_3 sources~~and of its precursorsto the~~ , and their precursors, to the total tropospheric O_3 concentration ~~in a certain~~ of a particular area, a more thorough analysis of the ~~model ability~~ ability of the model to reproduce the observed meteorological variables is beyond the scope of this paper.

3.2 Contribution of tagged precursor sources to the MDA8 O_3 ~~concentration~~mixing ratios

Figure 2 shows the spatial distributions of simulated monthly mean MDA8 values from tagged O_3 source regions and types ~~for late spring~~ throughout late spring in 2010. The receptor regions ~~are~~ shown were mainly influenced by the overseas combination of NAM, ASI, OCN, and RST sources that ~~together have a contribution varying from 25~~ combine to contribute from 23 % in the ~~ALP region (that include Po Valley , characterized as one of the most polluted areas in Europe);~~ Po Valley to up to 53.6 % in the UKI region (see Table S1). O_3 from RST (a 7.5 - 15 % contribution) is the main ~~overseas source . The~~ source from overseas. O_3 ~~coming~~ derived from oceanic sources ~~affects mostly the~~ mostly affects Atlantic coastal countries (up to a 16.1 % contribution in the UKI region), yet a small contribution of ~4-5 % ~~can be seen over the~~ was also observed within inland regions. ~~The long-range transported~~ Long-range transport of O_3 from Asia and North America contributes ~~from~~ significantly to total observed O_3 in Europe, accounting for 9.6 % of the total observed O_3 in ITA and up to ~22 % in UKI and SCA. After intercontinental transport, ~~the~~ O_3 produced ~~elsewhere in~~ within Europe is an important source of O_3 in ~~the~~ receptor regions, followed by O_3 coming from other types (LGT, BIO, and BMB). In general, for this time of the year, the contribution from the local sources to the total MDA8 O_3 ~~concentration in the receptor regions is~~ mixing ratio in receptor regions falls within a range from 8.5 % (SCA) to 21 % (RBU) (see Table S1). Emissions from local sources do not only affect local O_3 mixing ratios, but also impact O_3 levels of bordering countries due to strong horizontal pollution transport. In all receptor regions, ~~the~~ local anthropogenic sources have a lower contribution to MDA8 O_3 ~~concentration is dominated by~~ mixing ratios than the sum of O_3 ~~produced by remote anthropogenic precursors, consistent with previous work~~due to anthropogenic sources in other European source regions and long-range transport of ozone from intercontinental source regions. The contribution of intercontinental transport to the total MDA8 O_3 mixing ratio in Europe is consistent with previously reported results, i.e. Fiore et al. (2009) and ~~Li et al. (2016)~~Karamchandani et al. (2017), while this study allows us to identify which anthropogenic sources exert a strong influence on MDA8 O_3 predicted in different regions. Using observations, Danielsen (1968), Thouret et al. (2006) showed that the transport of O_3 from the stratosphere also contributes to tropospheric O_3. Here, ~~the~~ stratospheric O_3 contributes up to 7 ppb (12.5 % in SCA) to the total MDA8 O_3 ~~concentration, similar to~~ mixing ratio, which is a finding similar to that reported by Derwent et al. (2015). A similar tagged system ~~, but for~~ for predicting O_3 levels, using the CAM-Chem model (Butler et al., 2018), ~~also shows that the~~ has also shown that stratospheric O_3 ~~has a large contribution~~ significantly contributes to the total

tropospheric O_3 ~~concentration~~mixing ratio. The MOZART chemical mechanism used in this study does not explicitly treat ~~the~~ stratospheric chemistry; thus ~~the~~ surface stratospheric O_3 could be attributed to the vertical and horizontal transport of stratospheric O_3 coming from ~~the~~ boundary conditions. ~~The emitted local sources do not only affect local O_3 concentrations, but they also impact the O_3 levels of bordering countries due to a strong horizontal pollution transport.~~

During June-August 2010, Western Europe was mostly influenced by a high-pressure ~~systems~~ system centered over the Atlantic (see Fig. S1). In the upper troposphere, a ridge influenced the vertical atmospheric structure, especially over southern Europe. Therefore, these "usual summer conditions" favoured the intrusion of warm air coming from Africa and the ~~Arabic Saudi~~ Arabian peninsula and led to a warm and dry climate characterized by subsidence, stability, clear ~~sky and high solar radiationintensity~~skies and high-intensity solar radiation. Hence, the photochemical formation of O_3 ~~formation is enhanced, leading to a stronger contribution from~~ was enhanced, and influenced the stronger contribution of local emissions to the total ~~concentration~~ mixing ratio compared to the previous period examined. Figure 3 depicts the average MDA8 O_3 for June-August 2010. For most regions, we notice ~~an enhancement~~ that levels of O_3 produced from local sources ~~in June-August compared with April-May~~ from June–August compared with April–May were enhanced (Figure 2). Local sources can contribute to more than 20% ~~to~~ of the mean MDA8 O_3 ~~concentration~~ mixing ratio (from 14.6 % in SCA to ~~33.6 % in ALP~~35.7 % in the Po Valley, see Table S1)~~, showing that the~~ . This shows that local sources play a ~~stronger role in~~ strong role in the formation of O_3 ~~formation during~~ throughout the June-August period, as has been previously shown by Jiménez et al. (2006) and Querol et al. (2018). Compared with late spring, the relative contribution of ~~the~~ overseas sources decreased in summer, varying from ~~12.8~~ 10.9 % in the ~~ALP~~ Po Valley receptor region to 44.8 % in the UKI region in ~~July (see Figs. 2, 3 and~~ the month of July (Figs. 2 and 3; Table S1). We ~~notice a reduced~~ noticed that the spread of O_3 produced from European anthropogenic precursors over ~~the~~ bordering regions compared with late spring 2010 (~~see~~ Figs. 2 and 3). The increase in average temperature combined with stable atmospheric ~~condition~~ conditions lead to an enhancement of the biogenic NO emitted ~~in~~ into the atmosphere, especially in South-Eastern and Eastern Europe; thus, the BIO source type contributes up to ~9 ppb (13.2 % of MDA8 O_3) in the RBU receptor region (see Fig. 3). The vegetation fires that took place across Russia in July and August (Gilbert, 2010; Huijnen et al., 2012) as well as in Portugal and Spain (European Commission, 2011) lead to ~~a~~ increases in the contribution of O_3 coming from BMB of up to 29 ppb (16 %) in the RBU receptor region and up to 8.5 ppb (2.3 %) in the IBE receptor region. ~~The~~ BMB emissions contribute ~~domain wide more than 3.6 % (ALP), the most affected receptor regions being~~ domain-wide more than 3 % (Po Valley), with the greatest impacts modelled over RBU, IBA, SEE, SCA, and TCA. Another consequence of enhanced photochemical activity during ~~the summer season is the reduction~~ summer is that it reduces the influence of stratospheric O_3 ~~that contributes in general up to ~3-4% of total~~ from a domain-wide mean MDA8 O_3 ~~concentration at the surface.~~ mixing ratio of 4.4 ppb in the spring to 1.3 ppb in the summer (Figs. 2 and 3).

The decrease ~~of the~~ in photochemical activity in September 2010 is reflected in ~~the decrease of~~ decreases in total O_3 ~~concentration~~ mixing ratios compared with the summer ~~season~~of the same year, as well as in ~~the reduction of~~ a reduction associated with the local source contribution to the total O_3 ~~concentration (see~~ mixing ratio (Fig. 4). Thus, only in IBE, TCA, FRA, ~~ALP and RBU region the~~ Po Valley, the high Alps, and RBU regions were contribution of local sources to total MDA8 O_3 ~~is~~ higher than 20 % (Table S1). On the other hand, we ~~notice~~ noticed an increase in O_3 coming from ~~overseas anthropogenic~~

~~sources and~~ anthropogenic overseas sources and from lightning in autumn, stressing ~~the seasonal variation in~~ that seasonal variations exist within the outflow from other continents~~, and the~~ . There also is variation in the lifetime of O_3 which is ~~shorter~~ shortest during the summer ~~due to the enhanced photolytic sink~~as a result of enhanced photolytic activity.

Although we have seen that ~~long range~~ long-range transport plays a major role ~~to~~ in total O_3 ~~concentrations~~mixing ratios, the tagging technique helps to gain more ~~insights~~ insight into which region of the world dominates these ~~concentrations~~ mixing ratios in spring or autumn. In early fall, the ~~western~~ Western European receptor regions exhibit a slight increase ~~by~~ of 1.6 % ~~of~~ in O_3 ~~concentrations coming from North-America compared with the springseason, whilst~~ mixing ratios coming from North America compared with spring, while the contribution of O_3 ~~concentrations coming from the~~ mixing ratios coming from other overseas sources ~~to the total O_3~~ decreases. This could be linked to the prevailing westerly wind ~~,~~and the synoptic conditions seen during the first period of September, when the Azores High extended far to the east and north (Fig S1)~~, creating such conditions that direct~~ . This phenomenon creates conditions that are conducive to the transatlantic transport of American pollution ~~can be seen far east~~in the eastern direction. For example, in autumn periods within the RBU receptor region, ~~the~~ North-American and oceanic sources ~~contribute~~ account for up to 14.6 % in spring and 11.4 % in autumn ~~to~~ of the MDA8 O_3 ~~concentration~~mixing ratios.

Apart from local and other type sources, ~~the~~ NO_x emissions from shipping activities in the Atlantic Ocean combined with the oceanic O_3 from boundary conditions are an important source of O_3 that explains up to 16 % in late spring, 21% in summer and 12% in early autumn of the MDA8 O_3 ~~concentration~~ mixing ratio in the UKI, IBE, FRA, GEN, CEN~~and SCA. Our results are similar to those presented by Tagaris et al. (2017) and the references therein, and Mertens et al. (2018) who showed that OCN contributes up to 20 % of total O_3 in the North Atlantic.~~ , and SCA regions. Butler et al. (2018) showed that O_3 from oceanic sources ~~reach a minimum~~ reaches a minimum level in the North Atlantic Ocean during the summer, yet this study shows that in the UKI, IBE, FRA, GEN, CEN, and SCA receptor regions the oceanic O_3 ~~peaks its maximum contributions~~ contribution peak in the summer. This implies that the nearby shipping emissions have a greater ~~contribution in these~~ impact on oceanic bordering countries rather than oceanic O_3 from ~~the~~ boundary conditions. Furthermore, the NO_x emissions from shipping activities in the Mediterranean and Black Seas ~~contribute~~ account for up to 14 % in late spring, 19 % in summer and 11 % in early autumn of the MDA8 O_3 ~~concentration~~ mixing ratio predicted in the receptor regions situated along the shore of the Mediterranean Sea, such as IBE, ITA, SEE, ~~ALP~~ and FRA.

~~As can be seen from Figs. 2-4,~~ Our model results has shown that the highest MDA8 O_3 ~~concentrations are predicted~~ mixing ratios are predicted to occur over the Mediterranean ~~basin~~ Basin. This is due to the ~~several more~~ presence of favorable conditions for ~~its formation like~~ O_3 formation including the presence of small deposition sinks and intense photochemistry ~~.~~ (Figs. 2–4). Several studies, such as Safieddine et al. (2014), Tagaris et al. (2017), Mertens et al. (2018), Querol et al. (2018) and the references therein, have used source attribution methods to establish the origin of tropospheric O_3 observed over the Mediterranean Basin. The tagging technique used here shows that the O_3 from shipping activities in the Mediterranean and Black Seas (MBS) explains, on average, 15 % in late spring, 20 % in summer and 12 % in early autumn of total MDA8 O_3 predicted ~~in~~ to accumulate within the MBS receptor region~~, similar to~~ . These findings are similar to those of Aksoyoglu et al. (2016) that showed these emissions ~~contribute within a range of 10 to 20 % to~~ accounted for 10-20 % of the mean O_3 in the

Mediterranean in the summer of 2006. Moreover, Tagaris et al. (2017) ~~shows~~ has shown that shipping emissions explain up to 30 % the MDA8 O_3 simulated for July 2006 over the Mediterranean Sea. This study ~~shows~~ has shown that the shipping activities ~~contribute~~ likely accounted for up to 35 % ~~to~~ of the MDA8 O_3 near the Strait of Gibraltar (see Figure 5) during ~~thr~~ the April-September 2010 period. ~~The shipping emissions have the highest contribution to~~ Shipping emissions contribute most highly to total O_3 in the Western Basin of the Mediterranean Sea. Aside from shipping activities, the other European source regions have a localized contribution to total MDA8 O_3 predicted in the Mediterranean Sea. Thus, ITA, ALP, GEN source regions ~~contributes~~ contribute mostly to the central basin; IBE and FRA are main contributors in the western basin and SEE and TCA ~~in~~ predominantly contribute to the eastern basin. Natural sources contribute on average up to 10% of MDA8 O_3 in the western basin, and up to ~~approximatively~~ approximately 25 % of MDA8 O_3 in the eastern basin. The ~~long range~~ long-range of O_3 transport contributes up to 45 % along the North African shore and it exhibits a zonal pattern, with low ~~concentrations~~ mixing ratios occurring in the North and high ~~concentrations in the South~~ mixing ratios occurring south of the Mediterranean Sea, a trend mostly due to O_3 ~~concentrations~~ mixing ratios from NAF and RST sources.

3.3 Tagged ozone precursor contributions to exceedances of MDA8 target values – case study

As previously mentioned, the European Air Quality Directive (EU directive 2008/50/EC, 2008) has defined a target value of 120 μg m^{-3} for the MDA8 O_3 concentration, which can be exceeded up to 25 days per calendar year ~~averaged over three years~~(over a three-year span). In the following~~we call exceedances the~~ , we refer to values that surpass 120 μg m^{-3} ~~, and non-exceedances the~~ as exceedances, and values below 120 μg m^{-3} as non-exceedances. Figure S2 shows the spatial distribution of the number of exceedances observed and calculated throughout the April-September 2010 period for the AirBase rural stations. The observed MDA8 O_3 exceeds the target limits locally in Po Valley, Austria, ~~Germany and the coastal area~~ and Germany; in coastal areas of Portugal, Spain, France and Italy; and inland areas of Poland and ~~isolated in Poland , and~~ Slovakia. ~~Yet, the modeled~~ However, the modelled exceedances do not exhibit the same spatial pattern or intensity as observed values. Our use of tags allows for the identification of main source contributors to exceedances of modelled MDA8 O_3. Given the high number of stations that measure O_3, for simplicity, we will discuss the source contribution to the MDA8 O_3 exceedances only for the ~~ALP~~ Po Valley, high Alps, and GEN receptor regions.

Figure 6 exhibits the contribution of each tagged source and type to ~~the modeled~~ modelled MDA8 O_3 ~~as well as the~~ and observed MDA8 O_3 ~~, samples~~ value. Samples were, in all cases, taken at the location of the measurement stations, ~~during~~ throughout the April-September 2010 period. Figure 6 shows ~~averaged conditions during~~ the average conditions that occurred during the exceedance of the MDA8 O_3 target value, and also~~at times during which~~ , at times, occurred when the target value ~~is~~ was not exceeded. ~~In order to~~ To perform the source attribution for the observed values, we have scaled these values proportionally by the relative concentrations of each tagged O_3 tracer in our model output.

The relative contribution of emissions from different source regions to ~~the~~ modelled MDA8 O_3 and to ~~the~~ observed MDA8 O_3 ~~scaled by~~ values, after being scaled to account for the contribution of ~~modeled~~ modelled sources of O_3 types is generally similar for ~~both~~ Po Valley and GEN receptor regions (see Fig. 6). In the ~~ALP~~Po Valley, we can ~~clearly~~ pinpoint the main remote contributor ~~is~~ as being MBS (see Fig. 6), followed by GEN, and FRA, suggesting a dominant westerly and northerly air flow.

The recirculation of air masses in the Gulf of Genoa could accentuate the sea breeze and therefore more O_3 coming from NO_x ~~precursors associated to~~ associated with shipping activities in the Mediterranean will be transported to the coastal and inland station.

The high Alps receptor region is less influenced by ALP emissions than the Po Valley, and is more influenced by remote sources (see Fig. 6). The increased contribution of O_3 from CEN, ITA and FRA to both exceedance and non-exceedance days in the high Alps receptor region compared with the Po Valley receptor region highlights the impact of the transboundary transport of O_3 and its precursors . Furthermore, the contribution of stratospheric as well as long-range sources was generally 6 % higher in this receptor region than in the Po Valley receptor region.

In GEN, the main remote source regions are FRA and CEN during the exceedance days and FRA and UKI during non-exceedance days (Fig. 6). Opposite to ~~ALP~~Po Valley, in GEN the model predicts ~~less~~ fewer MDA8 O_3 exceedances days. Comparing the source contribution to both ~~modeled~~ modelled and observed exceedances days, we ~~notice that model underestimates the~~ noticed that the model underestimates O_3 ~~concentration associated to the long range~~ concentrations associated with long-range transport and natural sources~~and predicts more~~ . Further, the model predicted higher levels of O_3 from CEN and FRA than observed. Underestimation of long-range transported O_3 into the GEN region in our model could be ~~an explanation for~~ explained by the fact that the number of ~~modeled~~ modelled MDA8 O_3 exceedances in GEN is half of the observed number of exceedances (~~Figure~~ Fig. 6).

This kind of analysis can be applied to improve our knowledge of the origin of O_3 ~~precursor's origin~~ precursors and their contribution to MDA8 O_3 health metrics. Hence, by ~~the means of the~~ using this tagging technique, ~~the policy makers~~ policymakers can identify future ~~action~~ actions required to control the NO_x emissions at local and regional levels.

3.4 Tagged ozone precursor contributions to regulatory ozone metrics

In this section, we discuss the contribution of O_3 ~~concentrations~~ mixing ratios from diverse emissions sources and types to several ~~different metrics which quantify~~ metrics that quantify the O_3 ~~exposure~~ exposures of humans and ecosystems. From modelled hourly ~~concentrations~~ mixing ratios of tagged O_3 sources and types, we have calculated different O_3 metrics, ~~such as~~ including non-cumulative (mean, MDA8, and the ~~95th~~ 95^{th} percentile O_3) and cumulative (SOMO35, W126, and AOT40) metrics. We have chosen not to analyse the performance of the calculated cumulative metrics in comparison with ~~the measurements, following~~ measured values, as was done in previous work by Tong et al. (2009). Their work showed that the poor performance of the cumulative metrics is closely related to the sensitivity of these metrics to the threshold values or weighting factors.

Figure 7 and Table S2 ~~exhibit the percent~~ include the percentage of the contribution of different ~~emissions sources~~ sources of emissions and types to total O_3 as calculated ~~for~~ using health and vegetation metrics. The non-cumulative O_3 metrics employed in this study have ~~a similar pattern~~ displayed similar patterns for most of ~~receptor regions: the~~ the receptor regions. The contribution of local and European sources to the total O_3 ~~concentration is~~ mixing ratios have been low when we ~~apply~~ applied to mean O_3 metric and high when ~~we are looking at 95th percentile , emphasizing~~ using 95^{th} percentile metric. These findings emphasize the importance of O_3 produced by local and neighbouring sources to the high end of the O_3 ~~concentration~~ mixing ratio distribution.

Splitting the non-cumulative metrics into early (April-June) and late (July-September) simulation periods clearly illustrates that the European receptor regions are more prone to ~~being influenced by the~~ be influenced by intercontinental transport during the early period than ~~during~~ the late period. The ~~intercontinental~~ contribution of intercontinentally transported O_3 ~~contribution~~ to mean O_3 values in different receptor regions is higher during ~~thecarly~~ the early period and it spans between 22.8 % and 54.3 % ~~, and it contribute~~ of total O_3. In the late period it accounts for between 16 % and 48.9 % ~~during the late season~~ of total O_3 (see Fig. 7 and Table S2). Since ~~in this case the~~ O_3 associated with intercontinental transport ~~is coming~~ comes, in this case, solely from boundary conditions, ~~it implies that~~ errors in boundary conditions affect the predicted ~~concentration~~ mixing ratio of various chemical species, ~~consequently~~ and, consequently, the contribution of ~~the overseas sources to European~~ overseas sources of O_3 to levels observed in Europe O_3 (Tang et al., 2007; Giordano et al., 2015; Im et al., 2018).

The ~~lower~~ shorter lifespan of O_3 ~~lifetime~~ over remote ocean regions ~~in~~ throughout the warm season~~combined with the synoptic conditionslead to a decrease of the intercontinental~~ , combined with synoptic conditions, has led to decreased levels of intercontinentally transported O_3 to Europe. Thus, for most receptor regions, the O_3 coming from Asia and the rest of the world was reduced by more than half when compared with the cold period. The O_3 ~~concentration~~ mixing ratio from the stratosphere is, in general, 2.5 times higher in the cold season than in the warm season which is consistent ~~with Butler et al. (2018) study~~ with the findings of a study by Butler et al. (2018) which showed that the stratospheric O_3 ~~concentration~~ mixing ratio varies with altitude and its lifetime is influenced by season and latitude. The tagging technique also helps to quantify the impact of biogenic and biomass burning emissions of NO_x on tropospheric O_3. The impact of biogenic NO_x emissions ~~to the~~ on mean O_3 ~~concentrations spans~~ mixing ratios is between 3.3 % in ~~ALP~~ Po Valley and 5.9 % in TCA in the early season, while during the late season it ~~spans between 5.6 % in ALP~~ is between 5.4 % in Po Valley and 13.4 % in RBU. The biomass burning emissions ~~explain a range~~ account for variable percentages of mean O_3 ~~concentration~~ mixing ratios. These span between 1.6 % in ITA to 5.3 % in RBU during the early season, and between ~~3.9 % in ALP~~ 3.8 % in Po Valley and 16.3 % in RBU during the late season. ~~Most of the time, the natural~~ Natural sources do not usually vary greatly when different non-cumulative metrics are applied~~except for~~ . An exception would be for the biomass burning emissions on RBU during the late season. Thus, BMB in RBU contributes to 16.3 %, 17.6 % and 28.8 % ~~to~~ of the mean, MDA8 and ~~95th percentile~~95^{th} percentile, respectively.

Even though the SOMO35 and AOT40 metrics are not accumulated over the same ~~time~~ period (SOMO35 ~~–~~ is accumulated over the entire simulated period, and AOT40 ~~–~~ metric is accumulated over the May-July period) and ~~they~~ do not use same input data (daily MDA8 O_3 for SOMO35 vs daytime O_3 ~~concentration~~ mixing ratios for AOT40), since they are based on threshold exceedances and are designed to measure ~~the~~ exposure to high O_3 levels of humans (SOMO35) ~~or~~ and vegetation (AOT40), ~~they give us the possibility~~ there is a way to directly compare ~~them. As can be seen in Fig.~~ data from each metric type. As shown in Figure 7 and Table S2, the contribution of different ~~emissions sources and types to the~~ sources of emissions and types as a proportion of total SOMO35 and AOT40 metrics ~~is~~ are similar for most of the European receptor regions. Their spatial distribution (not shown) is also comparable, with minimum values over the UK, NW Europe and Scandinavia and maximum values over Italy, the Alps, south of Spain, east of Turkey and in the metropolitan area of Moscow, Russia~~, consistent with the previous studies of~~ . These results are consistent with previous studies performed by Aksoyoglu et al. (2014), and Anav et al. (2016). The overseas sources ~~have a similar contribution to the~~ contribute similarly to SOMO35 and AOT40 indexes (usually

less than 30 %) for most of the receptor regions used in this study. However, in UKI ~~,~~ the overseas sources ~~have a contribution of~~ account for 32 % ~~to~~ of AOT40 and 38 % ~~to~~ of SOMO35, and in SCA they ~~have a contribution of~~ contribute to ~22 % ~~to~~ of AOT40 and 30 % ~~to~~ of SOMO35~~, suggesting~~ . This suggests that these metrics are ~~sensitive~~ more sensitive with respect to the O_3 ~~concentration~~ mixing ratios from remote sources in areas having a low level of O_3 pollution. In the RBU receptor region, these indicators are sensitive to O_3 coming from biomass burning emissions (20 % ~~for~~ of SOMO35 and 24 % ~~for~~ of AOT40), whereas for the remaining receptor regions the contribution of natural sources to SOMO35 and AOT40 is similar. ~~The local sources contribute within~~ Local sources account for a range of ~12 % (SCA) ~~-~~ to ~38 % (GEN) ~~to these metrics, accentuating the~~ of these metrics. These data highlight the occurrence of increased O_3 production from local sources in comparison with northern European countries as well as large emissions of NO_x in the GEN source region. Since the difference ~~in~~ between AOT40 and SOMO35 is only a few ~~percent, regardless~~ percentage points, regardless of the receptor region, we ~~could~~ were able to conclude that they ~~are efficiently complementing each other mostly due to the use of a threshold~~ behave similarly, according to thresholds used to define these metrics.

The tagging method allows a better understanding of the main precursor sources responsible for exceedances of regulatory O_3 metrics. This information can help to inform further modelling studies aimed at investigating the effects of emission reduction strategies, and ultimately inform air quality policy. For example, in the ~~ALP region, which includes the Po Valley~~ Po Valley receptor region, the modelled AOT40 is up to ~~2.6~~ 3.4 times higher than the target limit given by EU legislation (on average ~~23560.8~~ 31218 ppb – hours). The observed and calculated AOT40 values depicted in Figure S3 ~~clearly~~ exhibit the exceedance of target limits in ~~ALP. The~~ Po Valley. O_3 coming from local sources ~~explain 33.6~~ can explain 35.0 % of this value (~~AOT40 up to 9163~~ an average of 10909 ppb – hours). After ~~the~~ local sources, the main European anthropogenic sources contributing to high ~~level of~~ AOT40 ~~in the ALP region are distinguished to be FRA (6.5~~ values in the Po Valley region are from FRA (6.6 %), GEN (~~7.6~~ 7 %) and MBS (~~7.9~~ 8.8 %) (Table S2). Generally, the O_3 ~~concentration~~ mixing ratio and its precursors transported from other anthropogenic European sources ~~to the ALP receptor regions explains ~39 %, the natural sources ~13 % and long range transport ~15~~ into the Po Valley receptor regions account for ~39.5 %, while natural sources accounts for ~12.3 % and long-range transport accounts for ~13.4 % of the remaining AOT40 ~~concentrations~~mixing ratios. Thus, to reach at least the target limit in the ~~ALP~~ Po Valley receptor region, considerable emission reductions ~~are still~~ will still be needed, not only on a local scale ~~,~~ but also on the European scale, especially ~~in~~ within the MBS, GER, and FRA source ~~region~~regions.

Figure 7 also shows the ~~percent contribution of different emissions regions and types~~ percentage that different types of emissions and emission regions contribute to the W126 index. Interestingly, for most of the receptor regions, ~~the~~ local NO_x anthropogenic emissions cause the largest response in W126 values compared with the other cumulative metrics used ~~here~~ in this study. Thus, ~~the~~ local NO_x ~~precursors explain~~ explains from 10.9 % (0.1 ppm – hours) in SCA to more than 40 % of W126 in GEN (45.9 %~~(~~; 2.48 ppm – hours~~), and ALP (43.5 %(7~~ , and Po Valley (45.4 %; 8.7 ppm – hours) ~~)~~ of W126 index values calculated for each region. The effect of European transported plumes is also enhanced ~~for~~ when using the W126 index compared with the other metrics for most of the downwind receptor regions. This behaviour is related to ~~the way in which this metrics has~~ how these metrics have been defined. Due to its sigmoidal weighted formulation, as discussed in Westenbarger and Frisvold (1995), and Lapina et al. (2014), W126 ~~takes into account~~ includes all daytime values rather than O_3 levels above a

certain threshold, as is done using SOMO35 and AOT40; therefore lower weighting factors of less than 0.5 are given to low O_3 values and weighting factors above 0.5 are given to O_3 values situated above the inflection point of 67 ppb. ~~Given that all daytime values are considered by W126 and they are not disregarded as done with SOMO35 and AOT40, we explain why W126 is more sensitive to local NO_x precursors than other metrics.~~

The ~~modeled~~ modelled mean AOT40 and W126 ~~in the ALP receptor region are exceeding the standards (23560~~ values in the Po Valley receptor region exceeded standards (26368 ppb − hours for AOT40 and ~~24.5~~ 28.9 ppm − hours for W126) during the May-July 2010 period~~) andas we have seen from~~ , and, as shown in Fig. 7 and Table S2, ~~the~~ local sources are an important contributor to these metrics. To better understand why the W126 index is mainly influenced by local sources compared with the other cumulative metrics, ~~a more thorough comparison between~~ we more thoroughly compared AOT40 and W126 ~~over the ALP receptor regionis presented in the following. Figure 8 presents the~~ values for the Po Valley receptor region. As shown in Figure 8, a temporal series of hourly daylight values for mean O_3, W126 and AOT40 values averaged over the ~~ALP receptor region . Given~~ Po Valley receptor region are given. Since that the W126 unit is ppm − hours, ~~for~~ a more direct comparison ~~,~~ with the W126 index ~~is~~ would require values be expressed in ppb − hours. ~~It can be noted that all metrics have a similar temporal variation, peaking in~~ Further, all metrics showed a similar level of temporal variation in which they peaked in the first half of July. Also, whenever the averaged O_3 ~~concentration is~~ mixing ratio was lower than 60 ppb (Fig. 8a), W126 ~~is~~ value was lower than AOT40 (Fig. 8d)~~,~~. This way of acting was most probably due to the weighting factor being less than 0.3, and above this ~~concentration~~ mixing ratio W126 tends to be higher than AOT40. This ~~behavior~~ behaviour is closely linked to the definition of these metrics. If the O_3 ~~concentration~~ mixing ratio is less than 40 ppb, W126 has a weighting factor lower than 0.03, while AOT40 has a weighting factor of 0. Above this threshold, AOT40 has a weighting factor of 1, while in the case of W126 only O_3 values higher than 100 ppb have a weighting factor of 1. Due to ~~the metrics definition~~way these metrics are defined, predicted O_3 values in each grid cell are accounted for the W126, ~~and~~ may not be accounted for the AOT40 index.

In addition, ~~the~~ visual analysis of the time series also revealed that when the O_3 ~~concentrations~~ mixing ratios from local sources are ~20 ppb, these ~~concentrations~~ mixing ratios have a higher contribution to W126 than ~~to~~ AOT40. To better understand this observation, we have further ~~analyzed~~ analysed the relationship between mean O_3 values from ALP sources (O_3-ALP) and the percent contribution of these O_3 tracers to mean O_3, W126, and AOT40 metrics. Figure 9 ~~presents the scatter plots of~~ shows scatter plots for O_3-ALP ~~and the contribution of these concentrations to~~ that relate the contributions of these mixing ratios on mean O_3, W126, and AOT40. In addition, the linear regression of Y vs X (Y=a*X+b) using all data sets ~~has~~ have been applied. ~~It can be seen~~ We saw that, in general, high mean O_3-ALP ~~concentrations contributes more~~ mixing ratios contribute more highly to W126 than to AOT40; this ~~is also confirmed by~~ was also confirmed when the highest slope (~~1.64) attained when the linear regression is~~ 1.52) was attained when at linear regression was applied to W126 vs. O_3-ALP. ~~The averaged~~ Averaged O_3-ALP and mean O_3 as well as O_3-ALP and W126 ~~are~~ were highly correlated (r=~~0.97, and respectively~~ 0.96, and r=0.93~~), whilst~~ ,respectively), while O_3-ALP and AOT40 ~~have a lower correlation~~ are correlated more loosely (0.88). The high ~~correlation of~~ level of correlation between O_3-ALP ~~with~~ and both mean O_3 and W126 could be related to the fact that these metrics account for all ~~modeled~~ modelled values, whilst AOT40 considers only O_3 values above 40 ppb.

Extending this analysis to all receptor regions, we can explain why the W126 index is more sensitive to O_3 coming from local sources compared with the other cumulative metrics. In addition, W126 accentuates the contribution of BIO and BMB in RBU, TCA and SEE, most likely because ~~this metric considers~~ the metric includes all daytime values, and not ~~only~~ just those above a certain threshold. Thus, the use of W126 highlights the considerable ~~impact~~ impacts of BIO and BMB emissions ~~which are important sources of~~ on total O_3 ~~during rhe summer seasons and vegetation fires~~ mixing ratios throughout the summer and from burning vegetation that ultimately influence the extent to which O_3 ~~damage on~~ causes damage to vegetation.

We have seen that the contribution of ~~different~~ NO_x ~~precursors~~ to total O_3 varies ~~with metrics and with the region~~depending on metrics and regions considered. Hence, the tagging method could help design different emission control strategies in specific source regions depending on which impacts need to be reduced in specific receptor regions.

4 Conclusions

Here, we implemented ~~into the WRF-Chem model~~ a new chemical mechanism within the WRF-Chem model to account for source attribution of O_3 from NO_x ~~precursors~~. We investigated the origin of surface O_3 using the "tagging" technique ~~during~~ from April-September 2010, as well as the contribution of different sources to O_3 metrics, and their exceedance events.

Using tagged simulation from WRF-Chem, we show that the spatial ~~distributions~~ distribution of simulated monthly mean MDA8 from tagged O_3 ~~sources~~ source regions and types ~~for~~ throughout late spring, summer and early autumn of 2010. The contribution of different sources to O_3 production varies with season. We have identified ~~the~~ intercontinental transported O_3 as an important contributor to the total O_3 ~~concentration~~mixing ratio, especially in the late spring ~~,~~ and early autumn~~, while during summer~~. During summer, however, the O_3 production is dominated by national and intra-European sources. We have also identified shipping activities in the Mediterranean Sea as an important source of O_3 for the IBE, ITA, SEE, ~~ALP~~ and FRA peripheral maritime receptor regions. We also analysed ~~which are~~ the main sources of MDA8 O_3 over the Mediterranean Basin and ~~our study has identified the main contributors to high~~ we have the main factors that contribute to MDA8 O_3 ~~concentration,~~ mixing ratios to the greatest degree. These were mainly shipping activities and the localized contribution from the bordering countries.

To ~~have a better understanding of~~ better understand the origin of MDA8 O_3 exceedances, we compared modelled and observed values of MDA8 O_3 concentration in the ~~Alps and Germany~~Po Valley, high Alps, Germany, and Benelux receptor regions. ~~Thus, we have seen that during the exceedances days~~ Throughout days exceeding the recommended thresholds of 120 μg, the contribution from local sources ~~sources is ~45 %~~was ~41 %, 34 % and 38 % of ~~modeled~~ modelled MDA8 O_3 for Po Valley, high Alps, and GEN, respectively. Throughout days not exceeding recommended thresholds, local emissions explain ~27 %, ~~whilst during non-exceedances values is ~32 %and 2-3% for ALP, respectively GEN~~16 % and 23% of modelled MDA8 O_3 for the Po Valley, high Alps, and GEN, respectively. Moreover, this tagging approach revealed that the main remote sources of MDA8 O_3 are MBS, GEN, and FRA for the ~~Alps~~ Po Valley receptor region, and are FRA, CEN and UKI for ~~the~~ Germany and Benelux receptor ~~regions~~region. In addition, ~~this~~ these analyses identified a persistently high contribution of

transboundary sources to ~~the~~ background O_3 concentration in the ~~ALP~~ high Alps receptor region. Furthermore, by showing that the contribution of precursor sources to modelled O_3 target value exceedances in the GEN region is systematically different ~~to~~ from the contribution of precursor sources to modelled O_3 when exceedances are observed but not modelled, we have identified a possible reason (underestimation of long-range transport) for the poor performance of our model ~~at~~ with respect to reproducing the observed number of O_3 target value exceedances in the GEN region.

~~By means of~~ Through comparisons with different O_3 metrics, we quantified the impact of local vs. ~~non local~~ non-local NO_x ~~precursors~~ on O_3 production for each European receptor region. The comparison between mean, MDA8 and ~~95th~~ 95^{th} percentile O_3 metrics accentuate the importance of large contributions from different NO_x ~~precursors~~ to the high-end of the O_3 distribution. By analysing these metrics for two periods (April-June and July-September), we can clearly distinguish the contribution of different NO_x ~~precursors~~ to total O_3 ~~concentration~~ mixing ratios in each region ~~for~~ and throughout different times of the year. When we compare the cumulative metrics, we ~~notice~~ noticed that the SOMO35 and AOT40 indexes exhibit ~~a~~ rather similar behaviour. Considering that these metrics are not calculated over the same ~~time~~ period nor do they use same input data, the similar behaviour is likely due to the similar threshold values applied to define these metrics.

The use of the W126 index accentuates the importance of local emissions. To confirm this, we investigated the behaviour of ~~modeled~~ modelled mean AOT40 and W126 ~~in the ALP~~ values in the Po Valley receptor region. We noticed that when the O_3 ~~concentrations~~ mixing ratios from local sources are approximately 20 ppb, these ~~concentrations~~ mixing ratios have a higher contribution to W126 than they do to AOT40 ~~,~~ and determined that the difference was mostly due to the definition of W126 which takes into account all O_3 values, not only those that are above a certain threshold.

Overall, this study has identified ~~the~~ local and remote ~~contribution~~ factors that contribute to the MDA8 O_3 ~~concentration~~ mixing ratio during several periods as well as ~~to~~ within different O_3 metrics. Furthermore, the method applied here could be used to design ~~different~~ improved emission control strategies depending on which impacts need to be reduced.

Appendix A

- chemics_init.F;
- module_input_chem_data.F;
- module_plumerise1.F and module_add_emiss_burn.F to account the source attribution of biomass burning emissions to O_3 concentration;
- module_emissions_anthropogenics.F to account for the impact of anthropogenic emissions on O_3 concentration;
- module_bioemi_megan2.F and module_data_mgn2mech.F to see the impact of biogenic emissions on O_3 concentration;
- module_lightning_nox_driver.F for lightning-generated nitrogen oxides

- Dry and wet deposition of tagged trace gases are treated by module_dep_simple.F and module_mozcart_wetscav.F, thus all tagged species have the same dry deposition velocities and wet removal rates with the corresponding non-tagged species;
- module_ftuv_driver.F to consider the photolytical reaction of the new packages;
- emissions_driver.F;
- chem_driver.F.

Code and data availability. The WRF-Chem model is publicly available on http://www2. mmm.ucar.edu/wrf/users/download/get_source.html. The modification introduced and described in Section 2 as well as the model data can be provided upon request to the corresponding author.

Author contributions. AL and TB designed the research. AL adapted the automatic mechanism-rewriting and code-generation tools and in implemented into WRF-Chem source code. AL performed the model runs and subsequent analysis. AL wrote the pape with contribution from TB.

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Table 1. List of tagged European source ~~/receptor~~ regions

Acronym	List of countries
MBS	Mediterranean, and Black Seas
BNS	Baltic, and North Seas
CEN	East Austria, Hungary, Czech Republic, Slovakia, Estonia, Latvia, Lithuania, Poland
ALP	West Austria, Switzerland, and North Italy ~~(including Po Valley)~~
ITA	South Italy, and Malta
SEE	Bulgaria, Romania, Moldavia, Albania, Slovenia, Croatia, Serbia, Montenegro, Macedonia, Greece, and Cyprus
IBE	Spain, and Portugal
UKI	United Kingdom, and Ireland
GEN	Belgium, Netherland, Luxembourg, and Germany
SCA	Finland, Norway, Sweden, Denmark, and Island
FRA	France
RBU	Russia, Belarus, and Ukraine
TCA	Turkey, Azerbaijan, Armenia, and Georgia

Table 2. Observed mean and simulation summary statistics for meteorological parameters. The normalized mean bias (NMB) and correlation coefficient (R) are calculated between simulated and observed meteorological observation from GWO during April – September 2010 period

Variable	Observed			~~Modeled~~ Modelled mean			NMB (%)	R
	min	mean	max	min	mean	max		
MSLP (hPa)	1000.96	1014.3	1022.06	969.05	1014.5	1039.03	-0	0.98
T2M (°C)	-17.14	14.99	32.10	-22.50	14.76	43.45	-3	0.91
WS10M (m/s)	0.36	3.37	10.83	0.00	3.59	20.41	8	0.65
WD10M (°)	0	190	360	31.91	216	318	13	0.47

Table 3. Observed mean and simulation summary statistics for MDA8 O_3 concentrations ($\mu g/m^{-3}$) at rural background sites. The normalized mean bias (NMB) and correlation coefficient (R) are calculated between simulated and observed O_3 concentrations from the AirBase dataset during April – September 2010 period.

Analyzed period	**Observed**			~~Modeled~~Modelled			**NMB (%)**	**R**
	min	mean	max	min	mean	max		
April	52.5	97.0	140.8	36.5	90.8	134.5	-6.3	0.58
May	41.0	87.9	143.0	28.0	83.2	124.6	-5.4	0.62
June	44.2	96.2	162.3	32.0	89.7	132.6	-6.8	0.71
July	43.8	97.0	178.2	26.0	90.8	147.7	-6.3	0.58
August	40.3	87.5	145.2	27.3	82.6	130.8	-5.6	0.65
September	33.4	77.5	135.4	26.5	81.1	129.6	4.6	0.63
Total	40.5	90.5	160.5	28.4	86.3	135.9	-5.2	0.69

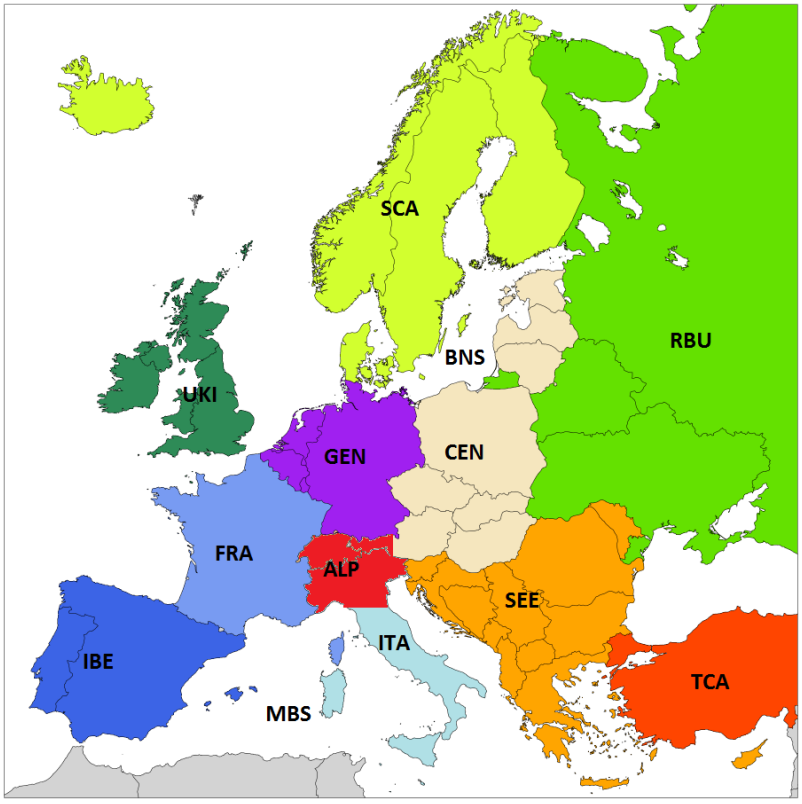

Figure 1. Tagged European source ~~/receptor~~ regions

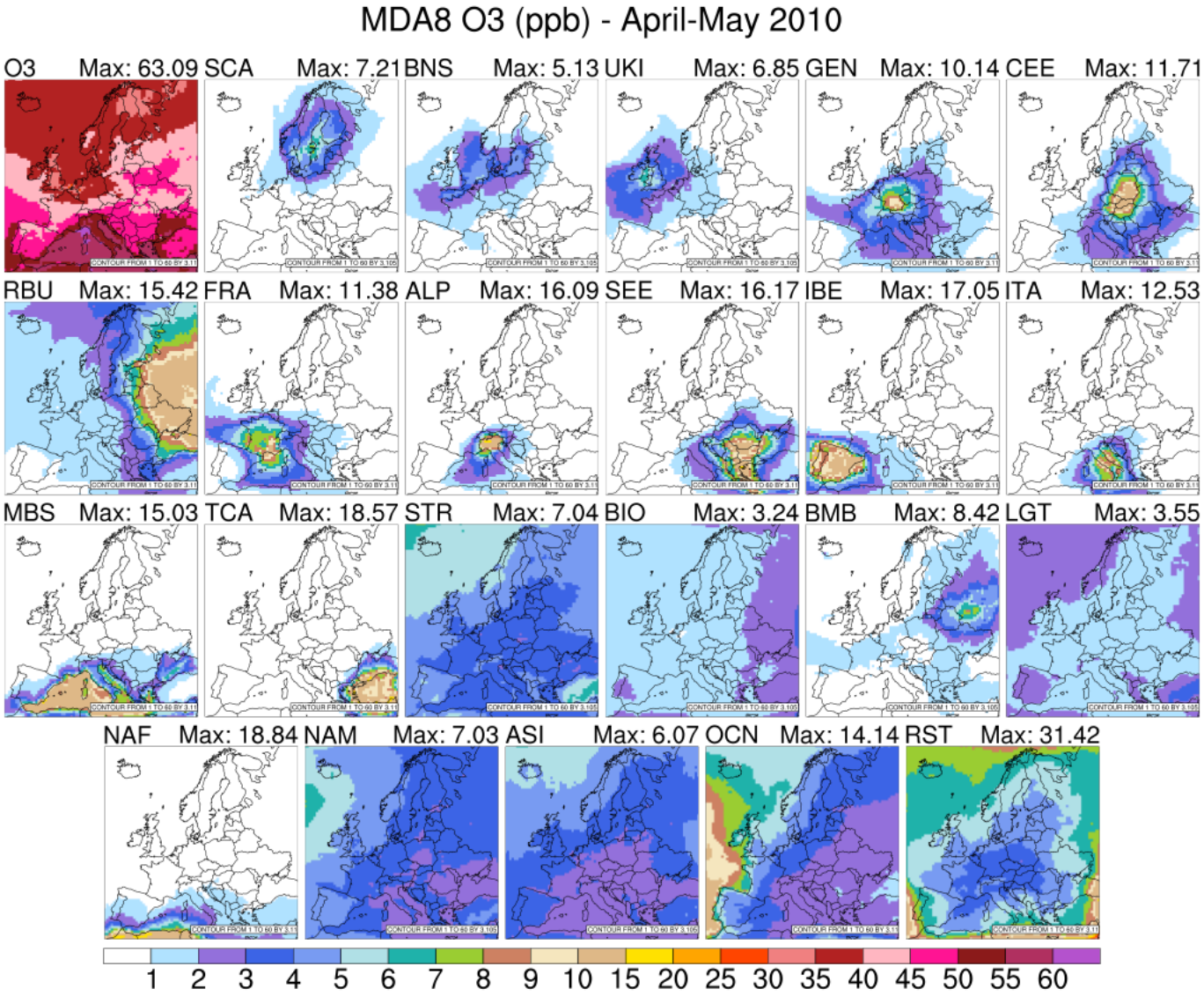

Figure 2. Contribution to MDA8 O_3 (ppb) of each O_3 source region for the April-May 2010 period

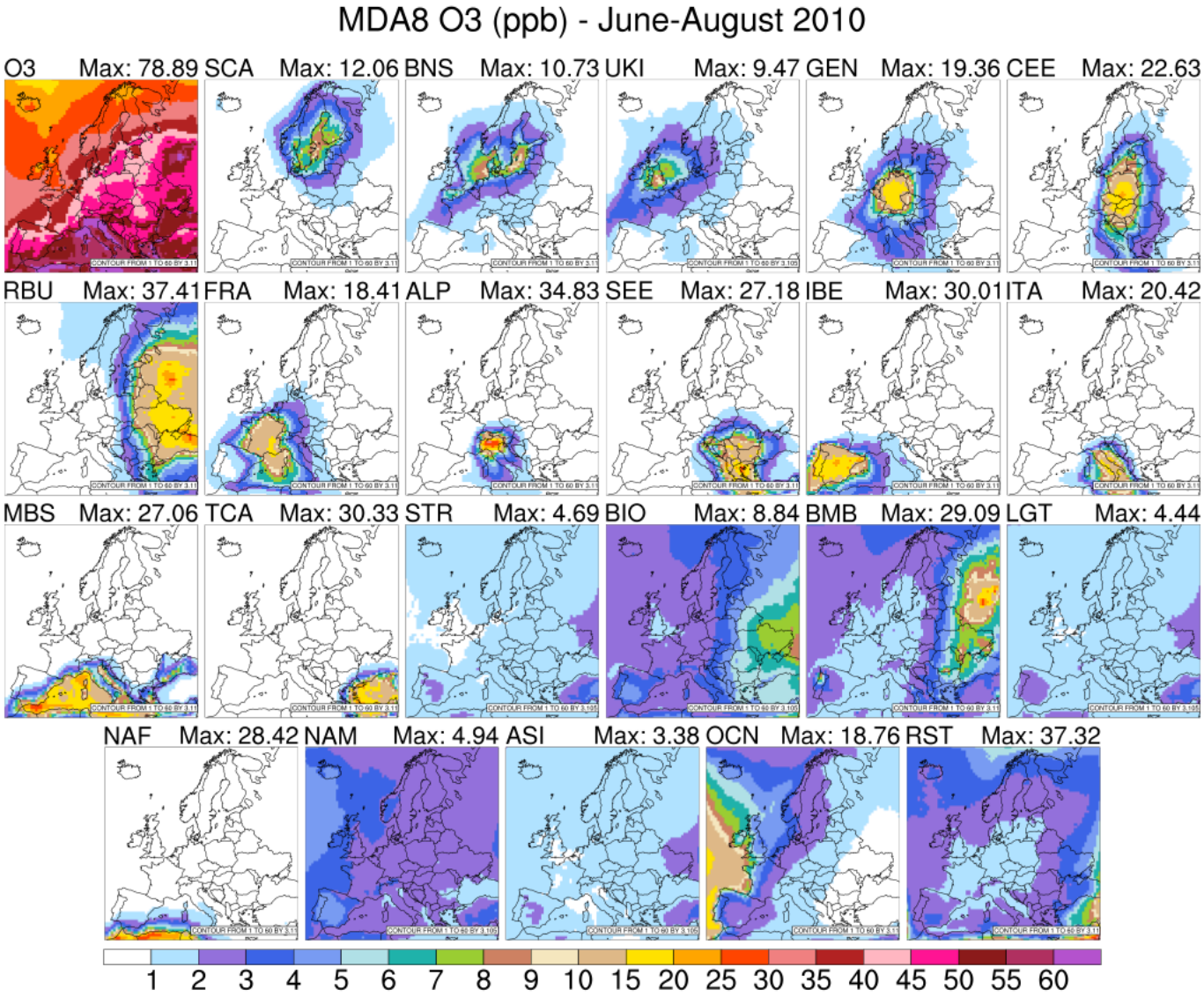

Figure 3. Same as Fig. 2, but for the June-August 2010 period

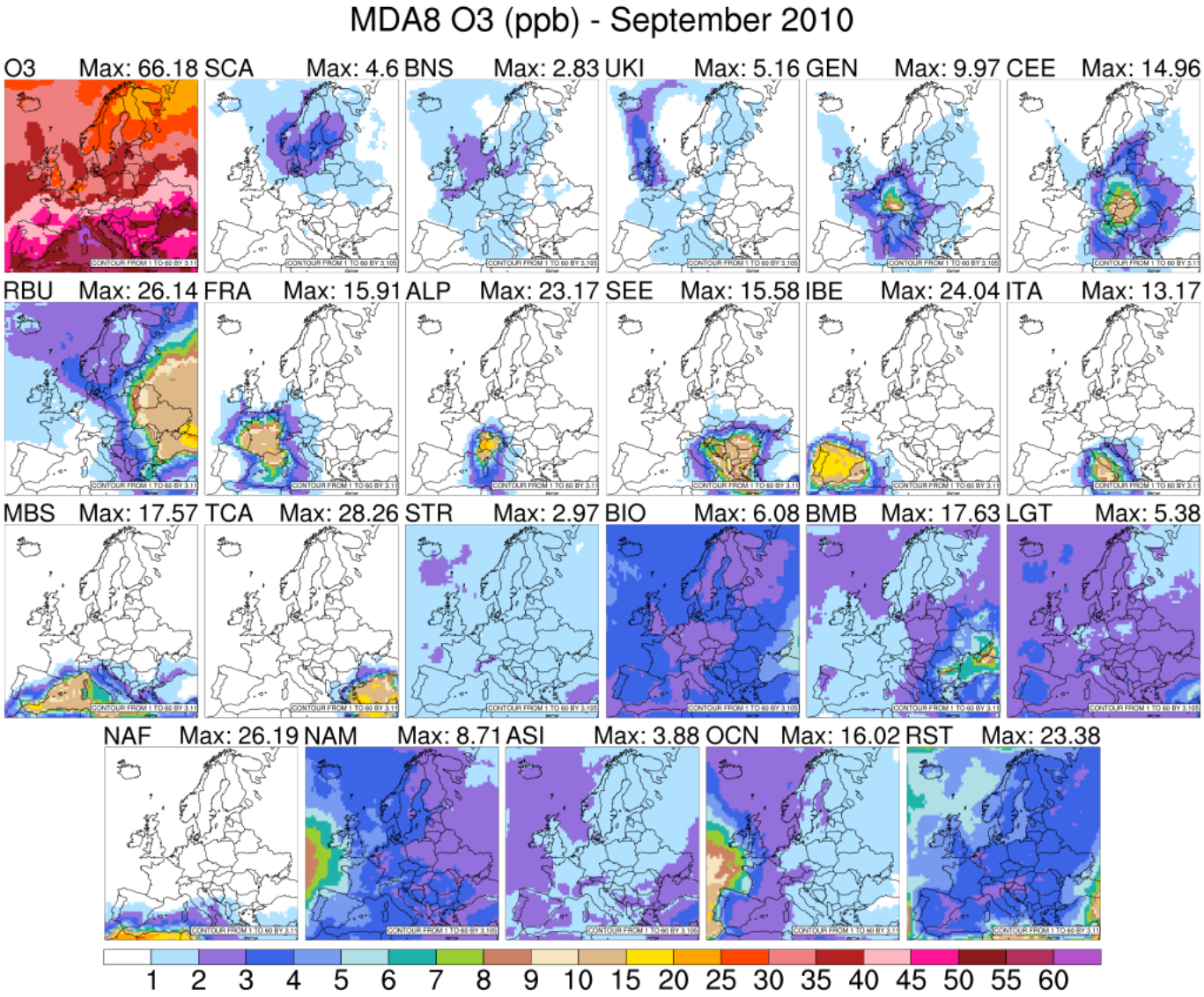

Figure 4. Same as Fig. 2, but for September 2010

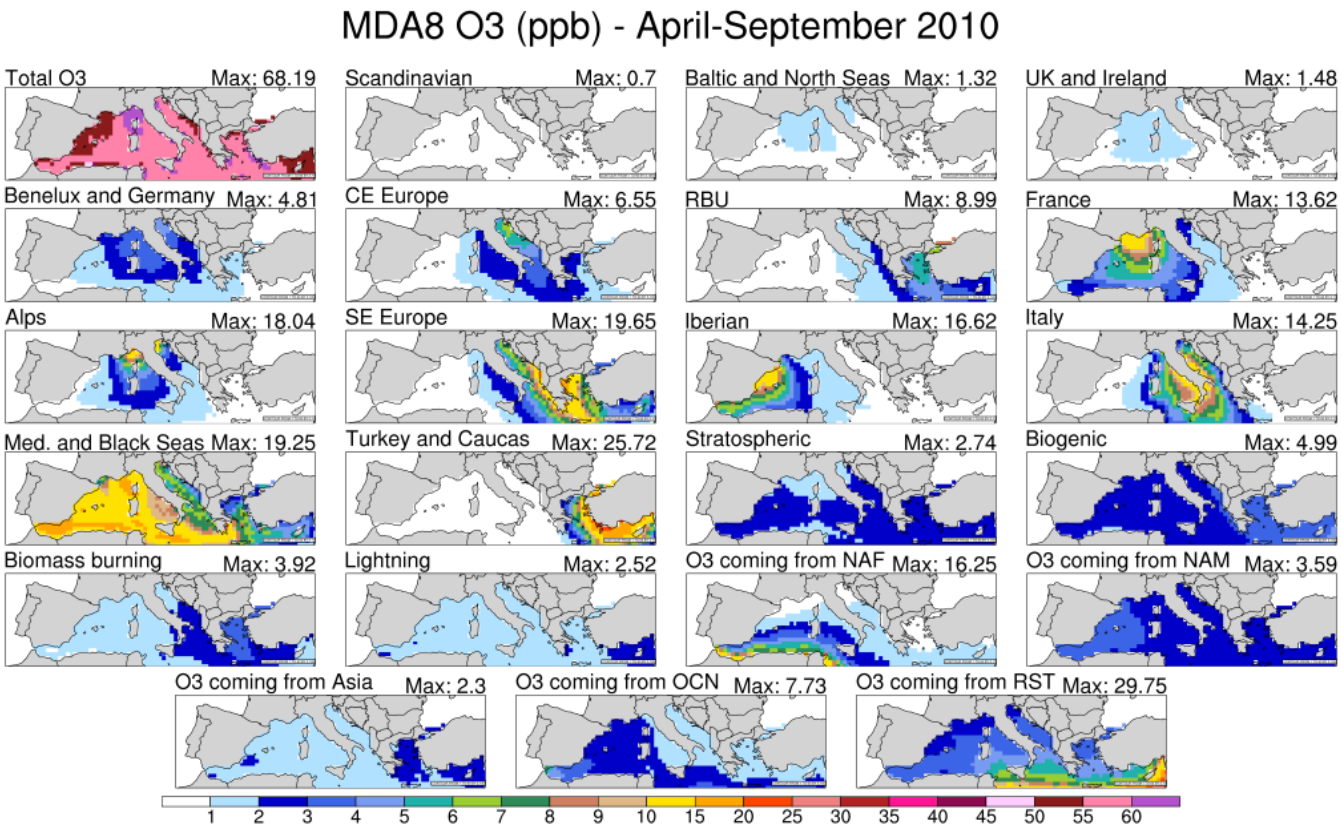

Figure 5. Average MDA8 O_3 mixing ratio (upper left panel) and contribution of each tagged O_3 region and source over the Mediterranean Sea for the April-September 2010 period. The unit is ~~ppb~~ppb

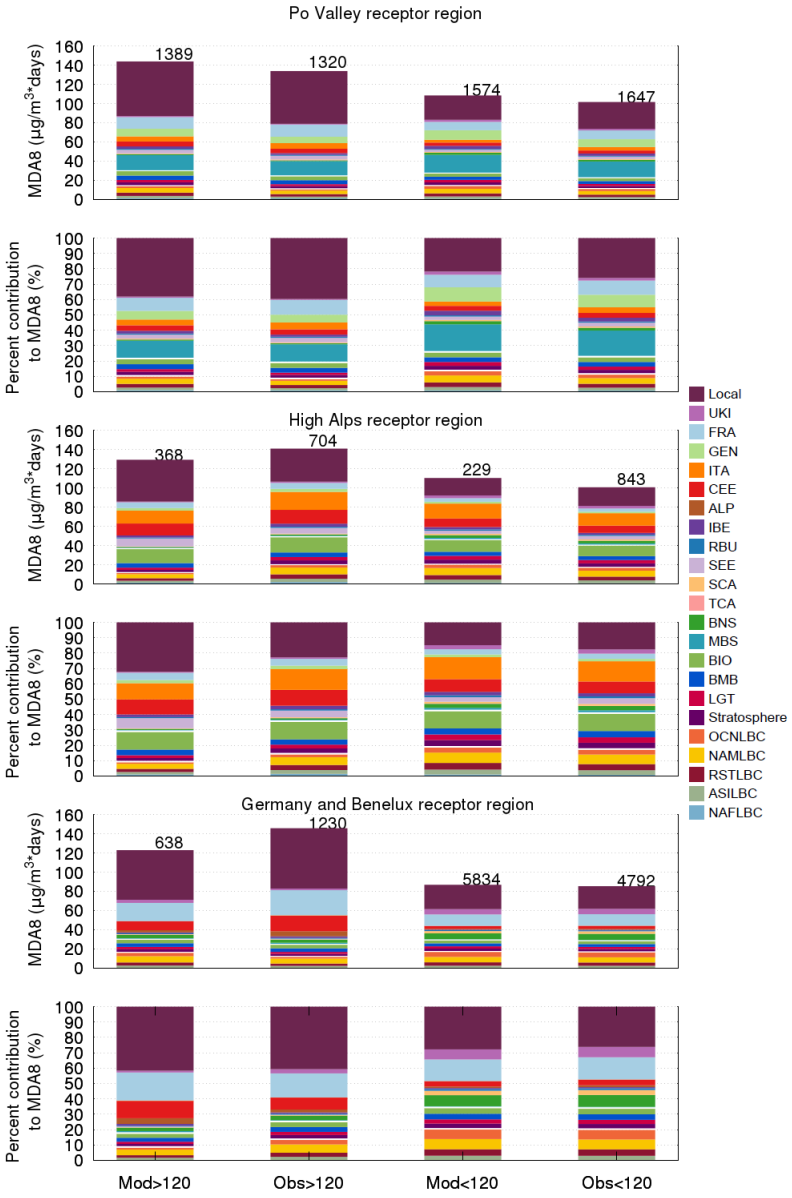

Figure 6. Mean ~~modeled~~ modelled and obseved MDA8 O_3 mixing ratio filtered by a threshold of 120 μ g m^{-}3 for ~~ALP~~ Po Valley, (top panel) high Alps (third from top panel) and GEN (~~second~~ fifth from top panel) and percent contribution to MDA8 O_3 from different emissions sources and types for ~~ALP~~ Po Valley (~~third from top~~ second panel), high Alps (fourth) and GEN (bottom panel) during April-September 2010 period. In each case the contributions of tagged sources to the total O_3 are shown. The tagged contributions to observed O_3 are obtained by scaling the observed O_3 by the relative contributions of these tagged sources to ~~modeled~~ modelled O_3. The total number of exceedances (and non-exceedances) of the MDA8 O_3 target value is indicated at the top of each column

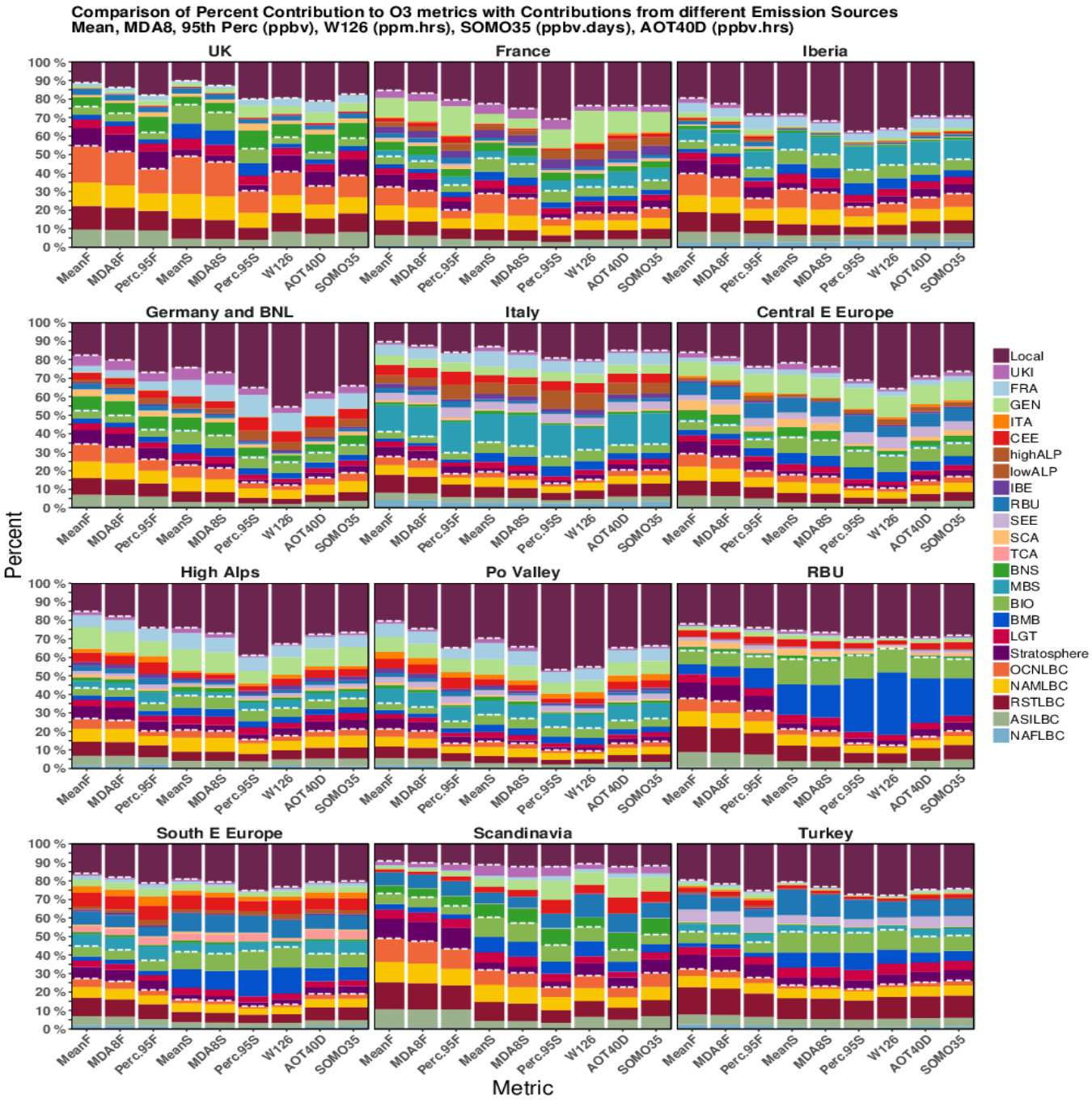

Figure 7. Comparison of percent contribution to O_3 metrics from different emissions sources and types. The metrics analysed are mean, MDA8 and ~~95th~~ 95^{th} percentile (ppb) for the early "F" and late "S" simulation period~~()~~, W126 ($\mathrm{ppm-hours}$), SOMO35 and AOT40 ($\mathrm{ppb-hours}$). The white dashed lines on each panel separate different categories (intercontinental transport, natural sources, and local and other European sources)

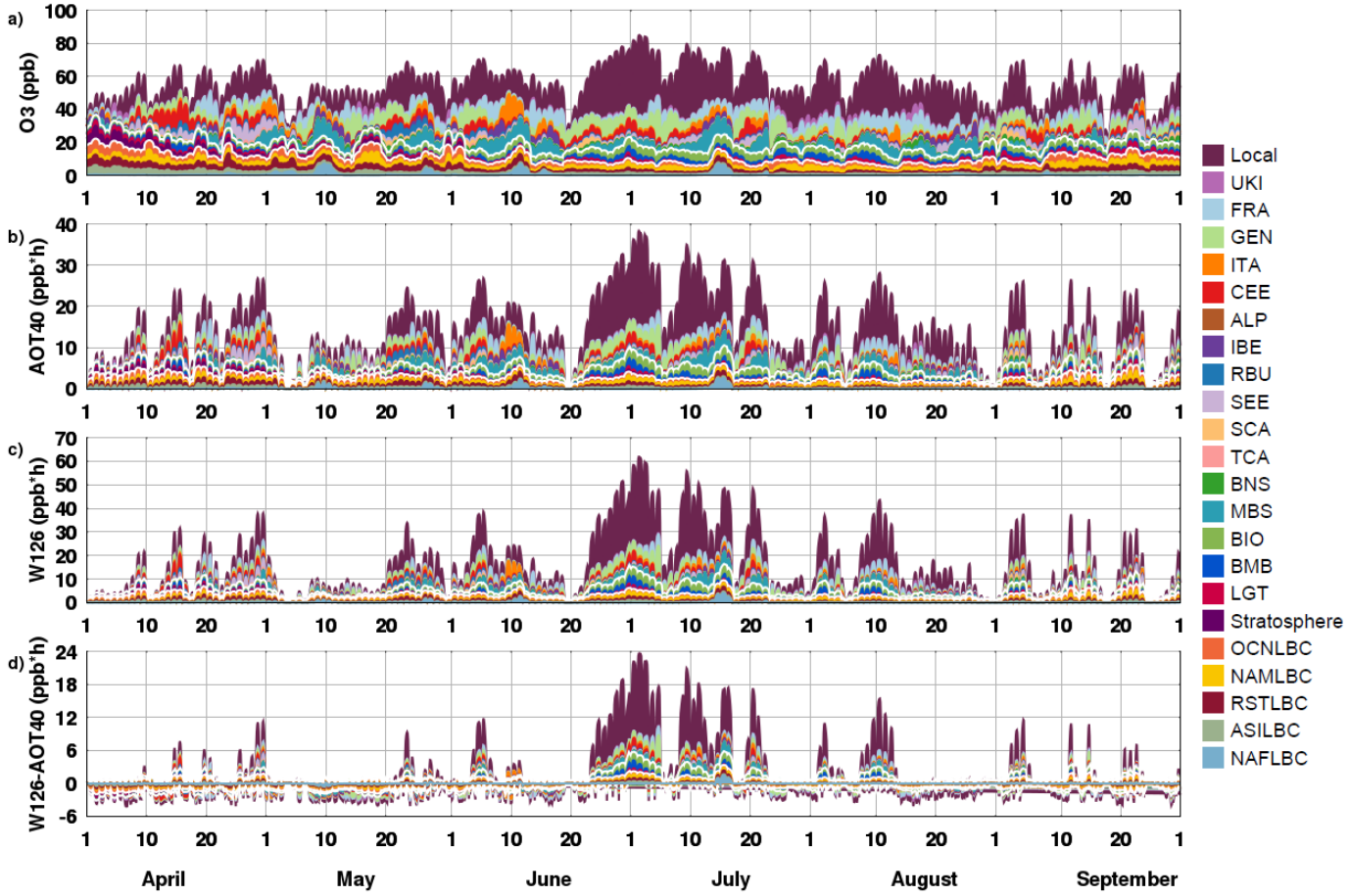

Figure 8. April-September 2010 time series of daytime a) hourly O_3 (ppb), b) hourly AOT40 index (ppb − hours), c) hourly ~~AOT40~~ W126 index (ppb − hours) and d) differences between W126 and AOT40 indexes (ppb − hours) averaged over ~~ALP~~ Po Valley receptor region

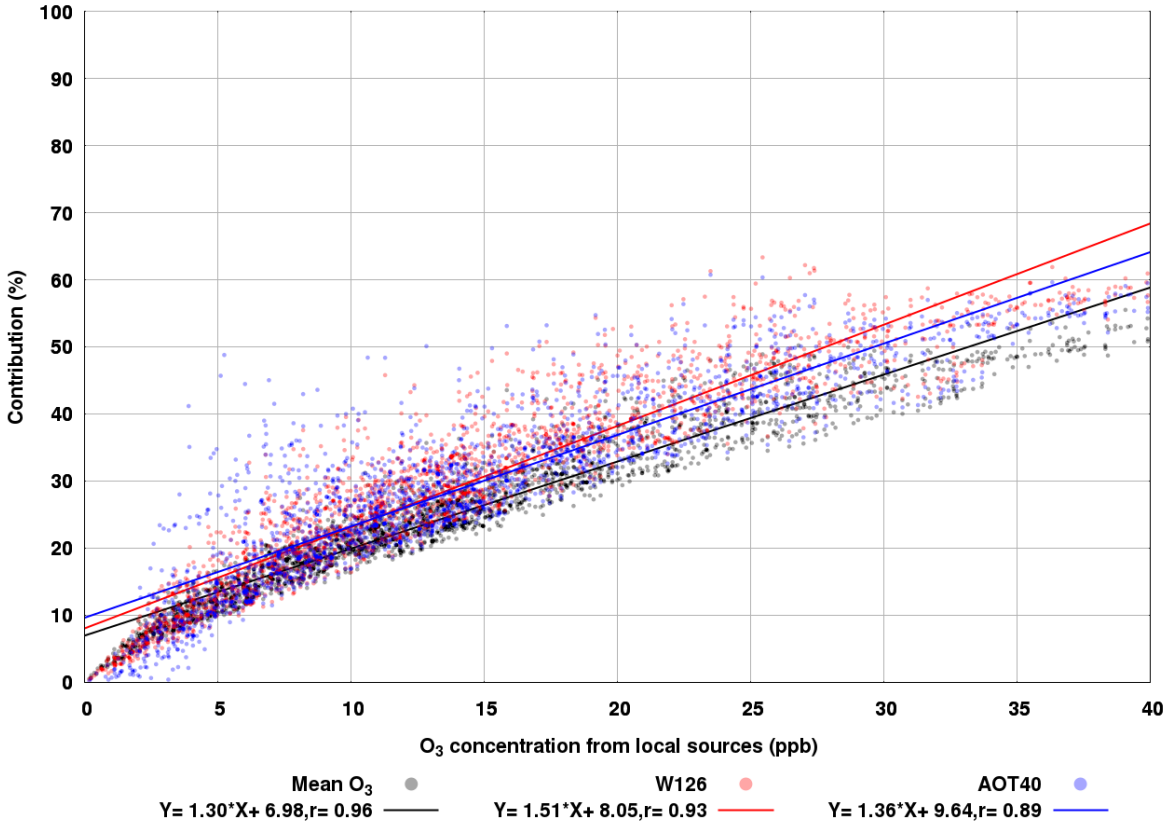

Figure 9. Scatter plots showing the ozone concentration from local sources versus the contribution to Mean O_3 (black dots), W126 (red dots) and AOT40 (blue dots). The solid lines are the lines of best fit.