#### **Response to Reviewers**

We thank the reviewers for their constructive and helpful suggestions; we have provided our responses to the reviewers' comments and believe our manuscript is much improved as a result.

### <u>Reviewer #1:</u> There are so many figures (13) and it could be condensed in approximately 7-8.

We reduced the initial number of figures (13) to 8 by deleting figures 8, 10 and 11; and by aggregating figures 2, 3 and 4, creating a new one (please see first figure attached). In addition, figure 12 (regression) was improved to include the bootstrapping results (please see second figure attached).

#### In line 11 of the abstract: CO2eq is not declared.

Thank you for your suggestion. We have now changed line 11 of the abstract to:

"...with carbon dioxide equivalent (CO<sub>2eq.</sub>) values..."

#### Introduction: Line 19-25: First paragraph should be at the end of introduction.

Thank you for your comment. We have now placed this paragraph at the end of the introduction.

## In Introduction, a paragraph with state-of-art in biomass burning emission is desired. Also, new methodologies that involve fire radiative power (FRP) and the ECMWF MACC system - which is running real time forecasts from FRP data to Europe, could be cited.

Thank you for your comment. This is a fair point. We have now included a new paragraph in the Introduction which addresses these issues.

"Most biomass burning emission studies rely on the model developed by Seiler and Crutzen (1980), which combines information on above-ground biomass available for burning, combustion factors, burning area, and emission factors for a certain species and vegetation type, to calculate the pyrogenic emissions (Wooster et al., 2005). However, these variables are hard to estimate, which causes large uncertainties in results. Recently introduced methodologies based on satellite measurements, are meant to improve these estimates (Wooster et al., 2004). Recent biomass burning studies both at regional (e.g. Pereira et al., 2009a) and global scales (e.g. Kaiser et al. 2009) rely on the relationship between fire radiative power (FRP), biomass consumed during a wildfire event, and smoke aerosol emission factor to provide estimates of the total amount of aerosol and trace gases emitted into the atmosphere (Woodster et al., 2005, Freeborn et al., 2008, and Pereira et al. 2009a). These new methods are especially important to overcome one of the main limitations of traditional methods: the difficulty in obtaining near real-time emissions estimates. Despite their advantages, these new methodologies still have uncertainties attached. The FRP satellite products, both from the Moderate Resolution Imaging Spectroradiometer (MODIS) and from the Geostationary Operational Environmental Satellites (GOES), require complex validations, yet unavailable. In addition, they also suffer from technical problems such as channel saturation, undetected fires, and cloud cover, among others (Pereira et al. 2009a)."

### The manuscript is well written but a few errors could be found in the text and in references, the most important are described below, but a detailed revision is required to final paper.

We modified the text and the reference list so that all reference problems were corrected:

- on page 22049, line 2 the reference was changed to "...(Brown and Smith, 2000)."
- on page 22049, line 9 the reference was changed to "...(Miranda et al., 2009a)."
- The references from Barbosa et al., 1999, Hodzic et al., 2007, and Korontzi et al., 2004 were added to the reference list. Furthermore, the studies from Effron (1982), Freeborn et al. (2008), Kaiser et al. (2009), Miranda et al., (2008), Pereira et al. (2009a), Wooster et al. (2004), and Wooster et al. (2005) were also added to the reference list. The two references from the same author and year (Fernandes et al., 2000) were corrected in the reference list to 2000a and 2000b.
- In addition, we have replaced the references from Fernandes et al. 2006, Oliveira 2008, and Tome et al. 2007, to the most updated versions of their studies, Fernandes et al. 2009, Oliveira et al. 2010, and National Forest Authority 2010.

Furthermore, we changed the text to correct small errors detected by the referee:

- on page 22049, line 21-22, the word "estimated" was deleted: "Their pyrogenic carbon dioxide..."
- on page 22050, line 12, we have deleted the Portuguese word "de": "... of 7.39 Mt CO<sub>2eq</sub>."
- on page 22051, line 11, we have replaced the word "prone" by "susceptible":"... the region susceptible to vegetation fires, ..."

As suggested by the reviewer, on page 22050, line 13, we have included a short description and declared  $CO_{2eq}$ .

"...Silva et al. (2006) estimated carbon dioxide equivalent ( $CO_{2eq.}$ ) emissions (...) in 1998.  $CO_{2eq.}$  is a universal measurement to evaluate the impact of releasing different greenhouse gases to the atmosphere. Each greenhouse gas (GHG) has a global warming potential (GWP), which estimates the impact of a chemical species in global warming, when compared to the impact of  $CO_2$  (which has a GWP of 1)."

Moreover, on page 22050, lines 19 to 21, we have rewritten the paragraph, as suggested by the referee.

"Pio et al. (2006) estimated pyrogenic dioxin emissions as the second largest source for this pollutant at the national level (17%)."

### On page 22050, line 22, referee #1 posted a question on what were the 3 factors considered by Schultz. The answer to this question can be found in the next sentence.

"Schultz et al. (2008) considered that three main factors of uncertainty limit the accuracy of longterm, global biomass burning emission data sets. These factors, also relevant at the regional scale of the present study are the <u>accuracy of estimates of burnt area</u>, <u>combustion completeness</u>, <u>and</u> <u>emission factors</u>."

## I think that Yokelson et al. (2008) should be referenced and maybe the difference in Emissions factor of Andreae and Merlet (2001) with this could be associated in uncertainty analysis.

We thank the reviewer for this comment; however, since the study from Yokelson et al. (2008) is only concerned with pyrogenic emissions from wildfires and pasture maintenance in tropical regions, we believe that the results presented by the author would not be appropriate to describe emissions from a wildfire in a southern European country.

## Why do not use emission factors derived in Zarate et al. (2000) in generalization between "Savanna and grassland" in agricultural emission?

We thank the reviewer for the suggestion; however, we believe that for methodological consistence it is better to use the same emission factors reference (Andreae and Merlet, 2001) for all chemical species. Future work will consider a wider range of emission factors and it will probably include not only the reference the reviewer suggested, but also emission factors available for Portugal (Miranda et al. 2009b).

## On page 22060, Line 1-7: The results showed in this paragraph isn't easily understood in figure 2. I think Figure 2 and 3 could be grouped in only one figure. A new figure could show the results of this paragraph. Also, what does means the class "burnt in 1990 to 2007 / 1990 to 1998 and 1998 – 2007"?

The results shown in the referred paragraph are only additional information provided to the reader, they are not meant to be shown on Fig. 2. Also, we believe that a new figure is not entirely necessary, since the text provides all the information.

On the other hand, we agree with the reviewer's comment stating that classes "Burnt in 1990 to 1999" and "Burnt in 1999 to 2008" are not easily understood. A clarification is needed. These classes include those areas that burnt more than once during the period under analysis. However, due to lack of information we assume that these areas are left to abandonment after fire, leading to fuel (shrubs and litter) accumulation. In the new figure (please see first figure attached), these classes were re-named to "Burnt more than once". In addition, a sentence was added to the text to be easier for the reader to understand what this class represents.

"In Fig. 2, the class "Burnt more than once" includes those areas that burnt at least twice during each period. Due to lack of information we assume that these areas are left to abandonment after fire, which leads to fuel (shrubs and litter) accumulation."

## In Figure 12, errors bars could be placed to indicate annual variability and statistical test must be performed such as Efron (1982) and student-t test.

This is a fair point. We addressed this issue, as suggested, by adding the results from the bootstrap error analysis to the regression figure (please see second figure attached). In addition, slope and coefficient of determination ( $R^2$ ) distributions results (mean ± s.d.) were added to the text.

"To understand the relationship between annual burnt area and GHG emissions, we established a linear regression (Fig. 7) forced through the origin, since absence of burnt area must correspond to no pyrogenic emissions. In addition, to assess the uncertainty in the regression we used a bootstrap method (Effron, 1982). A sample of 1,000 points was drawn to reconstruct the original curve and establish confidence intervals for the parameters (dashed lines in Fig. 7). The results revealed a slope distribution centred at 0.014 ( $\pm$  0.001, significant at 0.05 level, by Student's t-test) and a coefficient of determination ( $R^2$ ) distribution highly skewed to high values (0.95  $\pm$  0.03, significant at 0.05 level,

by Student's t-test), which indicates a good agreement between annual burnt area and annual pyrogenic GHG emissions."

## This methodology used four fundamental parameters well-known in literature but an accuracy assessment and inter-comparison is needed in context of emissions. This could be made using CO retrieved from Atmospheric Infrared Sounder (AIRS), SCIAMACHY or MOPITT.

Although we agree that a bottom-up (our results) / top-down (satellite measurements) comparison would help to assess the accuracy of our results, we do not see how this could be done in a rigorous quantitative way. Contrarily to what happens in tropical regions, CO emissions measured from satellite over a small European country like Portugal are a mixture of biomass burning as well as other sources (transports, industry, etc.). Since, as shown by this study, biomass burning CO emissions, represent only a small fraction of the total we would be comparing our results with a mixture of CO emissions from different sources.

In addition, CO emissions are quickly transported in the atmosphere to places far from where they have initially been emitted. Moreover, CO concentrations over Portugal measured by the satellite will also include emissions from different sources outside the country that quickly spread into the atmosphere.

For these reasons, we believe that a comparison between estimates from similar methodologies would be more sensible. Therefore, we compared our estimates with the official estimates that the Portuguese Environment Agency (APA) reports annually to the European Environment Agency (EEA). We added a paragraph to the discussion section (5.4) to address this issue.

"Our estimates are highly correlated (Pearson's correlation (r), all significant at 0.01 level) with the wildfires' GHG emissions reported by the European Environment Agency (EEA) for the same period in Portugal (Pereira et al., 2009b). The burnt area estimates are highly correlated (r = 0.95), as well as the GHG (r = 0.91),  $CO_2$  (r = 0.91),  $CH_4$  (r = 0.99) and N<sub>2</sub>O emissions (r = 0.99).

Both methods revealed 2003 and 2008 as the years with higher and lower emissions, respectively. However, our estimates are much more conservative (5,655 *versus* 11,289 Gg  $CO_{2eq.}$  in 2003), despite the fact that our burnt area estimates are higher (440,025 *versus* 286,000 ha in 2003). These differences arise mainly from significant discrepancies between  $CO_2$  pyrogenic emissions estimates. Contrarily to our study, EEA also account for indirect carbon losses, such as the natural decay of dead organic matter following fires, as well as harvesting emissions, which account for the loss of the entire dead tree at the time of fire (Pereira et al., 2009b). This can lead to an overestimation since it includes leaves, branches, wood, bark and roots. On the contrary, as mentioned above, we assume that in a typical fire only leaves and small branches are consumed, which can lead to some underestimation in the case of severe wildfires." Reviewer #2: The methods and scientific assumptions are not always clearly outlined; several times "grey literature" is referred as the basis for the applied methods. The authors should avoid to be based on this type of references only. If these are the only ones authors should develop more the description of the used methods, equations, and assumptions.

We agree with the reviewer as "grey literature" must be avoided as much as possible. Therefore, we replaced every possible "grey reference" with an international publication, when available. Unfortunately, in some cases this was not possible. However, in those cases an official and traceable report or document is always presented.

One conclusion is the possibility to estimate GHG emissions based on burnt area; I would like to have a stronger scientific basis for this conclusion (a simple linear regression is not enough). Anyway this correlation between GHG emissions and burnt area should be mentioned and stated (but not as a clear conclusion).

We thank the reviewer for the comment. The confidence in our results was improved by performing a bootstrap and error analysis on the linear regression between burnt area and pyrogenic emissions (please see our response to reviewer #1). We believe that the results from this analysis provide strong scientific basis to support the conclusion that knowing the annual burnt area is enough to have a good estimate of that year's pyrogenic GHG emissions. In the Abstract, we have replaced the word "accurate" by "good" in the sentence related to this matter.

## The traceability of results is not always possible. For instance authors don't provide neither mention the area burnt data source. Only the abstract contains this information and it is very generic.

Thank you for your comment. This is a fair point. We have provided further details on how the annual burnt area is obtained in the end of the "Burnt area" section (3.1).

"The fire perimeter atlas used in the present study is derived from Landsat satellite imagery for the period between 1990 and 2008, with a spatial resolution of 30m and a minimum mapped unit of 5ha (Pereira and Santos, 2003). The procedure is semi-automatic, starting with a supervised approach using classification trees, followed by on-screen editing of classification results. The final step – validation – is made by comparing the results against the official Portuguese field statistics (National Forest Authority, URL http://www.afn.min-agricultura.pt/portal/dudf/estatisticas) at the parish and county level."

The authors tried to do a comprehensive state-of-the-art, but one of the most important topics is not covered – new methodologies to estimate wildfire emissions. Also other wildfire emissions inventories and estimates are not entirely covered. Moreover, a comparison of those emission inventories data for wildfires in Portugal with this paper's data would be an added value.

Reviewer #1 as posted a similar comment; therefore, we added a new paragraph in the introduction with the new methodologies to estimated wildfire emissions. Please see the response to Reviewer #1 above.

In addition, as suggested by both reviewers we compared our results with other emissions inventories. In this case, we have compared our results with the official estimates provided by the European Environment Agency (EEA), which uses a similar methodology to estimate pyrogenic emissions. Please see the response to Reviewer #1 above.

The abstract provides a concise and complete summary of the paper. Only three remarks: line 9 – Monte Carlos is only mentioned here. Along the paper Monte Carlo is not clearly mentioned again. line 11 – clarify what is CO2eq. line 15 – "allowing for accurate emissions estimated" – do you really think these emissions estimate are going to be accurate?

We thank the reviewer for the suggestions. Regarding  $CO_{2eq.}$  please see above our reply to Reviewer #1 who posted the same comment.

We agree that Monte Carlo should be mentioned again in the appropriate section ("Uncertainty and sensitivity analysis", section 3.5). Therefore, we now included a sentence to state when Monte Carlo is used in our methodology.

"Model output variability was assessed through uncertainty analysis, followed by sensitivity analysis (Saltelli et al., 2004). Uncertainty analysis is based on a Monte Carlo approach. The first step in this procedure ..."

Regarding the last comment it was already addressed above.

1. Introduction The CO2eq concept is not explained at all. Page 22049 line 2- Brown et al or Brown and Smith? line 2 - 7 - this sentence is not clear to me. Please, revise it. line 10 - clarify Miranda et al, 2009: is it 2009a, 2009b, or both? line 11 - Hodzic et al is missing in the references list. Page 22050 line 11 - 19 - these sentences are not clear. Too many numbers (emissions, dates, percentages) are provided with too much detail. Try to rewrite them. line 27 - Korontzi et al is missing in the references list. Line 27 - 29 - you're saying that you used much higher spatial resolution satellite imagery and checked for accuracy against filed data, but you never describe what you used and how.

We thank the reviewer for the suggestions. As they are essentially the same as posted by Reviewer #1, we have already addressed them earlier in this document. Please see above.

Regarding the last comment we addressed it above.

### 2. Study area line 8 – Portuguese climate is temperate.

We have changed the word in the sentence from "Mediterranean" to "temperate".

"Climate is temperate, typically ... "

## <u>3. Data and Methods line 7 – you have to clarify which GHG are you considering. You're including other than only GHG gases in your analysis.</u>

In fact, we only clarify which GHG we considered in section 4.3. We agree that this clarification should be made earlier in the text. Therefore, we modified the sentence referred by the reviewer to address this issue.

"Calculation of GHG (carbon dioxide ( $CO_2$ ), nitrous oxide ( $N_2O$ ), and methane ( $CH_4$ )), other trace gases and aerosols pyrogenic emissions..."

<u>3.1 Burnt area The used land cover datasets are described, but how did you get the burnt area?</u> Based on satellite data? lines 11 - 12 - please explain how did you update the land cover mapswith the areas burnt annually. Table 1 - clarify what is "Burnt in 1990 to Burnt on 2007".lines 19 -20 - once again, describe how did you overlaid the land cover map with the fire perimeter atlas.What fire perimeter atlas?

Regarding the burnt area data we addressed this issue in the response to your comment above.

The procedure to update the land cover maps annually is a result of overlapping the annual burnt areas with the initial land-cover maps. Hence, we started each time period (1990 and 2000) with a land-cover map (COS90 and CORINE00, respectively). At each year, these maps were updated with the annual burnt area, simply by overlaying each land-cover map with the burnt area map using ArcGIS. This was done for both periods. Therefore, the land-cover map in 2005, for example, is the result of the overlay between the initial land-cover map (CORINE00) and the burnt area until then (2000-2005).

The class "Burnt in 1990 to Burnt in 2007" includes those areas that burn more than once during each period. So, if an area burn in 1995 and again in 1998 it will be included in this class. As we are considering two short periods (9 and 8 years, respectively) and we do not have enough information we assume that these areas are abandoned, which leads to fuel (shrubs and litter) accumulation. Since Reviewer #1 posted a similar comment, please see our response above.

### <u>3.2 Biomass estimation In this section you're using several references to support the description of</u> your methods, which are not peer-reviewed international publications. You should avoid this type of references to support your methodology description.

We strongly agree with the reviewer as not peer-reviewed international publications should be avoided. In fact, we avoided them as much as we could. Unfortunately, some of the information (like biomass equations) is only reported in official Portuguese documentation. We believe that, although not internationally published they provide the most accurate methods available to calculate biomass accumulation in Portuguese forests and shrublands.

We updated some of the references as newer versions of the documents are now available (5<sup>th</sup> National Forest Inventory, URL http://www.afn.min-agricultura.pt/portal/ifn/relatorio-final-ifn5-florestat-1) and the study from Oliveira (2008) is now submitted to an international journal (Oliveira et al., 2010).

## Table 2 – order the forest cover as in the previous table. Fernandes et al (2000b) is missing. At the references list you have to clarify what is Fernandes et al 2000a and Fernandes et al 2000b. Table 3 – Include BEHAVE in the table's caption.

We have corrected all the references and modified Table 3 to address all the comments.

### <u>3.3 Combustion factors You have to say that Table 4 data are coming from a bibliographic review.</u> <u>Table 4 – Pay attention to Fernandes et al 2000a; it not yet defined in the references list. Table 4</u> <u>doesn't include combustion factors for agriculture. So, what did you use?</u>

We modified the last sentence of section 3.3 to clarify that data are from a bibliographic review.

"Discrimination by major land cover type is unfeasible, due to lack of data. The data in Table 4 result from a review of the literature."

The lack of data on agricultural combustion factors is a limitation in our study. We did not find studies in the literature that address this issue. Therefore, we assumed that agriculture combustion

factors are within the same range of the forest combustion factors (Table 4) for the two biomass components (shrubs and litter) considered in the agriculture classes.

## <u>3.4 Emission factors line 16 – nitric oxide (NO) or nitrogen oxides (NOx)? Because when presenting and discussing results you mention NOx instead of NO.</u>

We thank the reviewer for noticing this inconsistence. In fact, we have estimated the nitrogen oxides  $(NO_x)$  emissions and not the nitric oxide (NO) emissions as stated by mistake in section 3.4. We corrected this error in the text.

"We calculated pyrogenic emissions of the following chemical species: carbon dioxide ( $CO_2$ ), carbon monoxide (CO), nitrous oxide ( $N_2O$ ), methane ( $CH_4$ ), nitrogen oxides ( $NO_x$ ), total nonmethane hydrocarbons (TNMH),..."

# <u>3.5 Uncertainty and sensitivity analysis Page 22057 line 23 – I don't understand why the variable space k is equal to 441. line 27 – the same applies to N equals to 256 and a total of 113408 runs. If you cannot easily explain why you're using these numbers, don't mention them. Page 22058 Line 15 – 18 – now you're mentioning Landsat-based annual burnt area maps and country-level field statistics. I really would like to have a better description of the used burnt area data at section 3.1.</u>

We agree that it is not very clear why N and k assume those values. The number of times the model is evaluated is given by the expression N(k+2), where N is the base sample (number of points) and k is the total number of variables (Saltelli *et al.*, 2004). We chose N=256 as compromise between having a reasonable number of points and computational time limitations. Regarding the total number of variables (or variable space, k), it is equal to 441 because we have to consider the annual burnt area in each land-cover class variables, as well as the biomass for each class (divided by biomass component) variables, the combustion factor by biomass component variables, and the emission factor by chemical species variables. In the end, summing all variables they are 441. Therefore, the model was run N(k+2)=256\*(441+2)=113,408 times.

We already addressed the burnt area comment earlier in this document.

### <u>I also would like to have a stronger basis for the 10% postulated coefficient of variation. Page</u> 22059 line 2 – equation 7 was not presented yet. The Olson's model is presented in equation 3. line 3 – What is Rambal data? line 7 – 8 – where are the combustion factors for agriculture? Or did I miss something?

The postulated 10% coefficient of variation as an uncertainty measure for the burnt area is based on a comparison between the Landsat-based estimates that we have used in our study with the official county-level field statistics of annual burnt area (National Forest Authority, URL http://www.afn.min-agricultura.pt/portal/dudf/estatisticas).

We changed the sentence to: "...Olson's model predictions (Eq. 3), assuming a normal...".

The question regarding the combustion factors for agriculture was addressed in the reviewer's previous comment. Please see above.

Rambal's model (Eq. 6) was used to estimate the leaf/biomass ratio, which was then used to estimate the litter accumulation using Olson's model (Eq. 5). However, Rambal (2001) does not provide any measure of error or variability of the model. Therefore, we would need the original data to see how the model was fitted and get an uncertainty measure. Since we lack that information we could not attribute any measure of uncertainty to the shrublands litter biomass accumulation variable, leaving these variables out of the uncertainty and sensitivity analysis.

<u>4. Results Figure 1 – take a look to the vertical axis; it is not starting at zero. Figure 2, 3 and 4 – Can't you join these 3 figures? Page 22061 Figure 6 – horizontal axis – kg instead of Kg. line 21 – how did you estimate CO2eq? Based on what GWP values? Figure 12 – include GHG in the figure caption.</u>

As stated above, we reduced the number of figures to 8. Please see above how we did it. We also corrected small errors in the other figures to address all the reviewers' comments (please see third figure attached).

Indeed,  $CO_{2eq.}$  were estimated based on global warming potentials for  $CH_4$  and  $N_2O$ . For clarification we added a sentence to the end of that paragraph to address this issue.

"Within the period under analysis GHG emissions reach maximum in 2003, with 5655 Gg  $CO_{2eq.}$  and minimum in 2008 with 159 Gg  $CO_{2eq.}$   $CO_{2eq.}$  estimates were obtained based on the GHG global warming potential (GWP) of CH<sub>4</sub> (GWP = 21) and N<sub>2</sub>O (GWP = 310)."

5. Discussion Page 22066 Line 26 – replace MP2.5 by PM2.5 Page 22067 line 17 – Miranda et al 2009b didn't publish emission estimates, but generic emission factors to be applied in Portugal. If you want wildfire emission estimates and their comparison with emissions from other sources take a look to Miranda et al., 2008 (Forest fires impact on air quality over Portugal. In NATO/CCMS International Technical Meeting on Air Pollution Modeling and its applications, 29, Aveiro, Portugal, 24-28 September 2007 – Air Pollution Modeling and Its Applications XIX. Springer: C. Borrego & A.I. Miranda, 2008, p. 190-198). Page 22068 Before starting analysing figure 13's content you have to mention the figure.

We replaced the reference (Miranda et al., 2009b) for the correct one (Miranda et al., 2008). In addition we corrected the two sentences mentioned by the reviewer.

"...variation (for example, PM<sub>2.5</sub>, OC and TNMH) revealed..."

"Miranda et al. (2008) published emission estimates for a range of land cover types..."

"Finally, it is interesting to compare vegetation fire emissions in Portugal with those produced by other GHG sources (Fig. 13) ..."

### <u>6. Conclusions line 9 – remember my comment regarding the gases for what you estimated</u> <u>emissions. Not only GHG were considered.</u>

We thank the reviewer for the comment. We re-wrote the sentence so it includes not only GHG but also the other trace gases and aerosols.

"We estimated national level, multi-year, pyrogenic emissions of greenhouse gases, other trace gases, and aerosols for Portugal..."

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