Authors Reply to Referee 1 (R1)

This manuscript presents a comprehensive study combining satellite information on wildfires and aerosol optical depth with trajectory modelling to illuminate the contribution of wildfire emissions on the European aerosol load. The manuscript is well written and organised and introduces significant new information. I have some comments and suggestions how to further improve this nice study. Altogether, I recommend publication after taking these minor modifications into account.

R1.1 - page 2318, line 1: no aitken model aerosol?

Approximately 80–90% of biomass burning aerosol volume is in the accumulation mode (e.g., Reid et al. 2005a), which is also the mode to which aerosol optical properties (as AOT) are most sensitive. However, following the Reviewer objection we rephrased the sentence as: *'Wildland fires represent the major source of fine aerosols (i.e., atmospheric particles with diameters < 1 µm)*', which is also more coherent with the terminology used throughout the text.

R1.2 - page 2318, line 3: 'the largest part' is an unclear phrase, because it may be related to the area burned or the amount of emissions produced

To avoid confusion we changed the sentence specifying 'the largest number of these fires occur ...'

R1.3 - page 2318, line 18: add 'the' Western Mediterranean **Done**

R1.4 - page 2319, line 6: add 'the' Russian Federation **Done**

R1.5 - page 2319, line 9: ignited instead of started **Done**

R1.6 - page 2319, line 25-38 and page 2334, line17-19: The weather conditions during April/May 2010 over Europe with the dispersion of volcanic ash from the eruption of the Icelandic volcano Eyjafjallajökull are an example for increased transport towards the south and east of Europe. Therefore I suggest to delete the example of the dispersion of the Chernobyl plume and maybe to better include a brief discussion of transport pathways dependent on NAO conditions.

The example of the dispersion of the Chernobyl plume has been removed. At the same time, we preferred not to introduce a further discussion in the text relating the transport pathways to NAO conditions since, according to results reported in the literature, this relationship is not straightforward. In fact, wind patterns favourable to transport from Eastern to Southern Europe may take place in positive NAO conditions (i.e., during enhanced advection of North Atlantic Air to northern Europe and following recirculation to south-western Europe (e.g., Trigo R., T. J. Osborn, J. M. Corte-Real, 2002, The North Atlantic Oscillation influence on Europe: climate impacts and associated physical mechanisms, Climate Research, 20, 9-17, 2002)) but may also occur in negative NAO periods, when the gradient between Azores High and Icelandic Low weakens the westerlies and allows the expansion of Siberian High toward Western Europe (e.g., Duncan, B. N., and I. Bey: A modelling study of the export pathways of pollution from Europe: Seasonal and interannual variations (1987-1997), J. Geophys. Res., 109, D08301, 2004).

R1.7 - page 2321, line 8: add fine particulate 'matter' **Done**

R1.8 - page 2321, line 25: did you check possible improvements of modifications published in Giglio et al., Biogeosciences 7, 1171–1186 (2010)?

We checked Giglio et al. (2010), but they present an improved "burned area" product, which is not used in our modeling framework. We employ MODIS Fire Counts and Fire Radiative Power data, which are not updated by Giglio et al. (2010). The use of burned area data could be an interesting extension of our study in the future.

R1.9 - page 2322, line 18-21: The sentence is a bit unclear to me: does it mean the MISR AOT evaluation against AERONET data is pretty good or not? And what does it mean for the study for sites with dust and smoke?

We meant to characterize MISR error following the current literature. At present, the Kahn et al. (2010) comparison with AERONET observations gives good agreement in the limits indicated in the text. To make the message clear, we revised the sentence as follows: '*The quality of the MISR aerosol products has been carefully evaluated by Kahn et al. (2010) through the comparison with relevant aerosol data from the worldwide AERONET sunphotometer network (Holben et al., 1998). That study shows good quality of the MISR AOT retrievals, with about three-fourth of MISR measurements within \pm 20\% of corresponding AERONET measurements, and about half within \pm 10\%. Focusing specifically on the 'biomass burning' category, the MISR mean AOT is well within the limits given above. Scenes with a large fraction of dust or smoke, generally display a poorer agreement'.*

R1.10 - page 2324, line 5: the chosen of resolution of 2.5_ is rather coarse, uncertainties due to this coarse resolution should be discussed in section 4.

In this revised version we specifically commented on this aspect in Section 4. Also note that our original sentence possibly led to a misunderstanding. We divided the domain into 2.5° cells and used these as starting point of the forward trajectories, but this is not the actual resolution of the Lagrangian model used to compute the trajectories themselves. This latter is only limited by the resolution of the meteorological data used, which in our case are NCEP analyses at 1° resolution. In the revised version of the manuscript we specified this aspect (Section 2.3) clarifying that: "Trajectories are driven by the 1°-resolution NCEP analyses. We divide the domain in a regular 2.5°- resolution grid and start trajectories from those grid-cell centres, where and when fires are detected. This resolution is chosen as a compromise between good spatial resolution of sources and acceptable computing time, because it limits the number of trajectories to be calculated."

R1.11 - page 2324, line 11: what is agl??

The acronym 'agl' is commonly used to indicate 'above ground level'. As not clear, in the revised version we now explicated the acronym.

R1.12 - page 2324, line 26: is an hourly resolution time step providing numerically stable results? Trajectories are actually computed with default HYSPLIT adaptive time step, which warrants numerical stability through the Courant-Friedrichs-Levy criterion. FWTD computations are then carried out considering trajectory positions at evenly spaced intervals of 1-hour, in order to avoid unbalanced summations among target points in eq. (1). To avoid confusion, we rephrased the related text in the paper as follows: "For each day of the month (starting month, sm), a 10-day forward trajectory Tn originating at the centre of cell n is computed and sampled at an hourly rate (provided that FCn > 25 fire count/1000km²/day)".

R1.13 - page 2325, line 18: in 2.5_ resolution, the European domain shown in Fig. 3 is subdivide into about 36 x 20 grid cells, however the figure suggests a much higher resolution. Please add more description if the FWTD has higher horizontal resolution or chose a more adequate and less smoothing algorithm for the illustrations.

As mentioned above, the 2.5° grid used for the starting points of the forward trajectories was chosen as a compromise between reasonable horizontal resolution of sources and computing

time. Nonetheless, coordinates (lat and lon) of the model-computed trajectories are given with 5 significant digits (see also our reply to your point R1.10 above). This means that, in principle, the FWTD fields can be obtained with a much higher resolution, dividing the target domain into smaller target cells (e.g., 1°-resolution cells as illustrated in the left figure below by the green grid). In this case 1°-resolution FWTD fields can be obtained, an example of which is shown in the right plot below (month of July, unsmoothed).



Note however that, for quantitative purposes, in our study we only use regionally-integrated FWTD fields (sum over q cells in Equation 4). Apart from some minor border effect, this regional integrals do not depend on the resolution of the target grid chosen. Therefore, to be consistent with the actual procedure we used based on a 2.5°-resolution target grid, we would prefer to leave Figure 3 (and Figure S1) as it is.

With respect to the colour smoothing in Figure 3 and S1, this was only done to improve the 'aesthetic quality' of the Figures. This can be changed if preferred by the Reviewer/Editor, leading to fields as the example below (July case, as the one above).



I would suggest not to focus only so much on the April fires, but at least to mention that the FWTD is maximum in August and that the whole continent is affected from the fires from July to September in particular in the east and the south.

Since in Figure 3 we presented the April and July cases as examples, we only commented these in this part of the text. Following the Reviewer's suggestion, we slightly changed the relevant sentence in the revised text.

R1.14 - page 2326, line 3: How many grid cells contribute to each of the seven selected European target areas?

In the 2.5° resolution grid used, the target areas are all composed of 28 cells, except for Scandinavia, which has 27 cells.

R1.15 - page 2326, line 16: instead of generally: expect in Western Europe and the Western Mediterranean

We rephrased the sentence as: "The figure confirms the expected maximum aerosol load in spring/summer and minimum in winter and further shows that in most of the addressed regions the AOT follows a clear bimodal yearly cycle, with maxima in April and July-August. This bimodality, particularly marked in the Central and Eastern European regions and less detectable in the Western ones, is also evident in the fine fraction AOT".

R1.16 - page 2326, line 17: instead of only July I would recommend to mention August as well, which is in several of the seven target regions the month with the second maximum **Done**

R1.17 - page 2326, line 17-19: I do not see the bimodality to be more evident in the fine fraction AOT in Figure 4. Please correct.

The sentence has been rephrased

R1.18 - page 2326, line 24: inter-annual instead of interannual Done

R1.19 - page 2326, line 25/26: Here the inter-annual variability is only mentioned to be also interesting. Please include some examples out of the period 2002-2007 which may be responsible for the extraordinary high or low AOD values in some of the seven target regions, e.g. variability in fire occurrences

Actually, the inter-annual variability in the period 2002-2007 is not only mentioned but it is also quantified in Figure 4 by the AOT standard deviations and commented in the text. To make this clearer, the sentence has been slightly modified in the text (see also answer to your next point R1.20).

R1.20 - page 2327, line 1-9: relatively unclear paragraph, please try to clarify, e.g. why should the summer minimum suggest the two maxima having non-negligible inter-annual variability?

At that point of the manuscript, we are commenting the standard deviation values (shaded areas in Figure 4) associated to the monthly mean AOT. Over the period 2002-2007, a low standard deviation associated to the monthly AOT indicates a reduced year-by-year AOT variability in that month. Conversely, the standard deviation increase in correspondence with the two AOT maxima further indicates these to be associated to phenomena having a non negligible inter-annual variability.

We rephrased this sentence as follows: "The inter-annual variability associated to both the AOT and FFAOT monthly means can be evaluated through the relevant standard deviations shown in Figure 4 (shaded areas indicate \pm 1 standard deviation). This variability is also interesting, and likely related to the irregular nature of fires. In fact, standard deviations show two minima, the first in the winter months and the second at mid-year (typically in May-June), in correspondence to the AOT minimum. In these months, such a reduced AOT variability over the period 2002-2007 indicates rather similar aerosol loads year by year. Conversely, the standard deviation increase corresponding to the two maxima further indicates these to be associated to phenomena having a non negligible inter-annual variability."

R1.21 - page 2327, line 22-24: it is mentioned that R>=0.8 in each region, please provide R for each region

Done

R1.22 - page 2329, line 9: please mention other 'disturbances' with pronounced seasonal cycles, e.g. soil dust and pollen mobilisation during harvesting and planting, biogenic aerosol formation during summer etc.

Done

R1.23 - page 2331, line 19: please correct: Stohl Done

R1.24 - page 2332, line 1: please take also into account intercontinental transport in higher altitudes from e.g. Asia, e.g. Lelieveld et al., Science 298, 794-799 (2002)

This interesting study by Lelieveld et al. 2002 reports results of a field campaign that took place in Crete in August 2002. Although in that case episodic transport from Asia was detected in the upper troposphere, that study well summarize how "In summer, a strong east-west surface pressure difference (> 20 hPa) is generated between the Azorean high and Asian monsoon low-pressure regimes. These quasi-permanent weather systems cause northerly flow in the lower troposphere over the Mediterranean, feeding into the trade winds further south. The flow is strongest and most persistent in August. In the free troposphere, on the other hand, <u>westerly wind prevails</u>. In the upper troposphere over the eastern Mediterranean, the flow is additionally influenced by the extended anticyclone centered over the Tibetan Plateau". This perfectly fits with the westerly flow we addressed in our work as the main driver of intercontinental pollution transport to Europe.

R1.25 - page 2332, line 11-14: it is unclear why it is sufficient to fit only the January-March and August to December values

As indicated in the manuscript (Section 3.1), this is done because the April-to-July period is affected by aerosol transport from America. Such period cannot thus be assumed as 'background conditions' as necessary for the fit.

R1.26 - page 2332, line 17 and 22, 23 and page 2333, line 1, 2: Fig. 9 is unnecessary, as the monthly mean wind field in 925 hPa does not tell much about transport pathways of pollutant plumes. The actual wind speed and direction and height of the plume determines the transport pathways. Therefore figures 8 and 9 do not present a prove for the aerosol production and transport from North-Central America.

Figures 8 and 9 address climatological properties of AOD and winds. Figure 8 shows the 2002-2007 monthly mean FFAOT (April and July) as derived from MISR. It therefore does prove the presence and the spatial gradient of aerosols off the North-Central American coasts, particularly in July. Figure 9 is the mean wind field in the same months at 925 hPa, an altitude representative of the free troposphere over the ocean, i.e., a likely altitude of plume transport. As such, it certainly does not represent a prove this aerosol is transported towards Europe. This kind of climatological maps are commonly used in the intercontinental transport studies (e.g. Auvray and Bey, 2005; Li et al., 2002). In our case, we believe Figure 8 and 9 provide the reader with a useful, climatological, picture of the two main factors driving the results of the study (i.e., the aerosol load and the wind field).

R1.27 - page 2333, line 3: see for BC measurements at Mace Head at the Atlantic Coast of Ireland for the variability on intercontinental transport of polluted air masses across the Atlantic

We checked for consistency with studies on transatlantic transport of BC at Mace Head, and actually found an interesting study that is now added to our discussion (Cooke et al., JGR 1997). It reinforces our expectation of a small contribution to the AOT from transatlantic transport, because air masses arriving from the Atlantic "clean" sector are found to be recently influenced by human activity only on 5% of the time.

R1.28 - page 2333, line 8-11: I suspect this is no misinterpretation as aerosol, but it is marine organic aerosol produced from phytoplankton, see e.g. O'Dowd et al., Nature 431, 676-680, 2004.

Thank you for this suggestion. In the revised version, we added this hypothesis into the relevant text.

R1.29 - page 2336, line 5, 6: isn't there a new directive only allowing the threshold of 50 microgram/m3 not to be exceeded more than 7 days per year

Yes, but it is not yet enforced as originally planned by the European Commission.