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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Wildfire particulate matter in Europe during summer 2003: meso-scale modeling of smoke emissions, transport and radiative effects" *by* A. Hodzic et al.

A. Hodzic et al.

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Manuscript reference ID: 1680-7375/acpd/2007-7-4705 "Wildfire particulate matter in Europe during summer 2003: meso-scale modeling of smoke emissions, transport and radiative effects" by A. Hodzic et al.

The authors are thankful to both reviewers for their suggestions. According to their reports, both reviewers are in favor of publication of this paper in Atmospheric Chemistry and Physics after minor revisions that have been addressed in this revised version of the manuscript.

1. SPECIFIC ANSWER TO REVIEWER #1:

Answer to Reviewer's General Comment:

The reviewer's major criticism concerns the novelty of the present paper, relative to our previous study (Hodzic et al., 2006, ACP). We agree that the objectives and new work performed in the present study need to be pointed out more clearly in the manuscript. Although the satellite measurements and the model used in the present study remain the same, the objectives and results are entirely independent from the previous one.

Hodzic et al., 2006 (ACP) evaluated the performance of an air quality model in simulating aerosols using optical data from satellites such as MODIS and POLDER during summer 2003, when the first set of data in cloud free conditions became available. A new comparison methodology was established and major discrepancies and uncertainties between model simulations and satellite observations were discussed and quantified. The model-observation discrepancies noted during the summer 2003 heat-wave episode were suggestive of long-range transport of wildfire emissions, but these effects were not quantified. Such emissions may become increasingly frequent in a warmer climate, but are not accounted for in current air quality models and air quality forecasting in Europe.

Therefore, in the continuity of this work, the major goal of the present paper was to add this capacity of simulating wildfires to the model and to evaluate impacts on local and regional air quality. The model was improved to take into account the MODIS daily smoke emission inventory as well as the injection altitude of smoke particles. The simulated aerosol optical properties were also put into a radiative transfer model to estimate (off-line) the effects of smoke particles on photolysis rates and atmospheric radiative forcing.

Although Hodzic et al. (2006) motivated the implementation of wildfire parameterizations in the model and the assessment of its impact in the present study, the new manuscripts contains a substantial amount of new results and should be considered as an independent study. This is now explained more clearly in the revised version of

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the manuscript. Specifically, the following paragraph has been added:

“Hodzic et al. (2006a) reported large inconsistencies in predicted aerosol concentrations and optical properties over Europe during summer 2003 caused by unaccounted emissions from wildfires. The present study extends this work by developing a new modeling framework that includes wildfire emissions and their effect on air quality. The objective of the paper is twofold: First, a simple parameterization of smoke emission and transport is presented and evaluated with observations; Second, the effects of smoke emissions on air quality in Europe are examined during the summer 2003 fire season, including both the direct impact on ground concentration of pollutants and the indirect impact on photolysis rates and atmospheric radiative forcing.”

Minor comments:

1) Pg 4706, Ln 17, add the year 2003 to (3-8, August) for completion. Done.

2) Pg 4706, Ln 15-22 rather than make a general statement about the model performance, you need to specify (as in the text) the extent to which the model simulations and observed AOT agree by using standard statistical methods such as root mean square error (rmse), or mean bias error. This would lend a better weight to the model’s efficiency. You may also want to mention that although the model generally reproduces the spatial dispersion of the plumes, the temporal variability of AOT data at specific AERONET locations is not well captured by the model.

Answer: The following paragraph has been changed in the new manuscript: “Although there was a fairly good spatial agreement with satellite data (correlation coefficients ranging from 0.4 to 0.9), the temporal variability of AOT data at specific AERONET locations was not well captured by the model. Statistical analyses of model-simulated AOT data at AERONET ground stations showed a significant decrease in the model biases and suggest that wildfire emissions are responsible for a 30% enhancement in mean AOT values during the heat-wave episode.”

3) Pg 4706 Ln 23, replace “First, directly” by “Firstly, directly” “First” sounds better that “Firstly” in this sentence.

4) Pg 4706 Ln 26, replace “Second, indirectly” by “Secondly, indirectly,” ”Second” was preferred to “secondly”.

According to previous remarks, the following changes have been made to the manuscript:

“First, directly, the modeled wildfire emissions caused an increase in average PM2.5 ground concentrations from 20 to 200%. The largest enhancement in PM2.5 concentrations stayed, however, confined within a 200 km area around the fire source locations and reached up to 40 ug/m3. Second, indirectly, the presence of elevated smoke layers over Europe significantly altered atmospheric radiative properties: the model results imply a 10 to 30% decrease in photolysis rates and an increase in atmospheric radiative forcing of 10-35 W/m2 during the period of strong fire influence throughout a large part of Europe.”

5) Pg 4707 Ln1 Can you briefly substantiate the lower or upper limit in the radiative forcing by indicating what aerosol property or meteorological condition is mainly at play. Answer: I do not understand this reviewer’s comment.

6) P4707, line 11-15, Provide the source for your estimate, otherwise change “according to our estimates generated” in Ln 12 to “according to our estimates could have generated”. Done.

7) Pg 4710, Ln 11, Include “atmospheric radiative forcing” as another indirect consequence. Done.

8) Pg 4713: Ln 6, AERONET data products are classified according to levels based on quality assurance. In addition to stating that the AERONET data used in this work are corrected for cloud contamination, you should also indicate the level of AERONET data products used.

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Answer: We use Level-2 AERONET data. The following was changed in the paper: “For this study, we use level-2 optical thickness data at 532 nm and Ångstrom exponent coefficients derived from the 440 and 670 nm channels.”

9) Pg 4717, Ln 22, Change “in the free troposphere within” to “in the free troposphere than within”. Answer: The correct sentence is: “transport of particles is more efficient in stable free tropospheric layers that are characterized by stronger winds.”

10) Pg 4720, Ln 17, Change “(referred as the)” to “(referred to as the)”. Done.

11) Pg 4721, Ln 7, Rather than wait until page 4722 to define “summer” as being used in this paper, you should rather do so earlier on (particularly on page 4721 where the word was mentioned but undefined.

Answer: The following was changed in the paper: “In this section, we first present the smoke emission estimates and the smoke optical signature over Europe from 1 July to 30 September, 2003, henceforth summer 2003 (section 4.1).”

“Figure 2 shows the distribution of total fire emissions as estimated with the MODIS satellite for the summer time period.”

“Figure 4 shows the Ångstrom exponents (alpha) and aerosol optical thicknesses measured at 532nm for the summer period at eight AERONET sites located respectively close to the main fire source region in the south-western Iberian Peninsula (Evora, El Arenosillo), in the Central and Eastern Mediterranean basin (Avignon, Oristano, Lampedusa, Rome), and in Northern Europe (Fontainebleau, Lille).”

And the figure caption was corrected:

“Fig. 1: Wildfire locations and estimated total fine aerosol (PM2.5) emissions (Tons=106g) derived from MODIS data over Europe for (a) 1 July to 30 September 2003 period, and (b) the heat-wave episode, 1 August to 15 August 2003. The map also represents the CHIMERE model domain.”

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12) Page 4721, Ln 22, Page 4722, Ln 5, You need to state whether Fig 2 is a 3-month composite plot or whether the data is averaged out. There is a wide margin between the scale of emissions on Fig 2 in comparison to Fig 3. Why is that?

Answer: Fig. 2 represents total PM2.5 smoke emissions (a) accumulated over a 3-month period, and (b) accumulated over the 15-day period. The values range from 0 to 15 kTons in each model grid; While Fig. 3 represents spatially averaged smoke emissions (respectively over Europe and Portugal) for each day in August and September 2003. Values range from 1 to 25 kTons for each day. The emissions values are numerically similar between two representations, although Fig.2 is plotted in Tons while Fig.3 is plotted in kTons.

The following clarifications have been added in the manuscript:

"Figure 2 shows the distribution of total fire emissions as detected by the MODIS satellite for the summer time period."

"Fig. 2: Wildfire locations and estimated total fine aerosol (PM2.5) emissions (Tons=106g) derived from MODIS data over Europe for (a) 1 July to 30 September 2003 period, and (b) the heat-wave episode, 1 August to 15 August 2003."

"Fig. 3: Time series of PM2.5 wildfire emission estimates derived from MODIS data and spatially averaged over Europe (total area) and Portugal (shaded area) for (a) August and (b) September 2003 months. Emissions from wildfires exceeded the total European anthropogenic emissions (EMEP database) during 3 extreme fire events that occurred in Portugal on 4-5 August, 11-14 August and 12-13 September, respectively."

13) Page 4722, Delete Fig. 3 from Ln 10, since your claim that "wildfires emitted 130 kTons of primary) is not verifiable from Fig 3. Fig. 3 only presents estimates of PM2.5 wildfire emissions for August and September and not July. Rather, add "Fig. 3" to "Table 2" on Ln 14, where it is more appropriate. Done.

14) Pg 4723, As claimed by the authors, low Ångstrom exponents denote the pres-

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ence of large particles. Specifically the low Ångstrom exponents reported here were attributed to dust particles. Other than the low Ångstrom exponents, do the authors have any other way of verifying this claim (e.g. visual observation of dust by trained observers, satellite imagery, chemical trace study e.t.c.), since low Ångstrom exponents in some cases could also be indicative of aged smoke. In addition the authors should consider adding the aerosol single scattering albedo to the upper panels of Fig. 4. While aerosol optical depth reflects the aerosol burden and Ångstrom exponent provides information on the aerosol size, the aerosol single scattering albedo indicates whether an aerosol type is scattering or absorbing. Since biomass burning aerosols are mainly absorbing, the easiest way to show their presence and distinguish them from other aerosol types (such as dust) is to depict the aerosol single scattering (see for example Iziomon and Lohmann, 2003). In addition, in discussing their results the authors should be reminded of the possibility of a mixture of dust and combustion aerosols as indicated by VanCuren [2003], who report that Asian dust is mixed with a substantial amount of combustion aerosols. Perry et al. [1999] also observe that black carbon is frequently mixed with dust over Hawaii in the springtime.

Answer: The dependency of the Angstrom exponent on the size distribution is commonly used (e.g. Eck et al., 2003; Reid et al., 1999) to distinguish between various aerosol components (desert dust, urban emissions, biomass burning particles Ě). In our study, the identification of dust particles is based on Angstrom exponent, as well as the results reported in literature and back-trajectory analysis. We agree that it can be useful to consider also the SSA, especially in case of a mixture of dust and combustion aerosols mentioned by the reviewer. Therefore, the following explanations have been added in the manuscript:

"The single scattering albedo (SSA) data are also considered in order to determine aerosol capacity to absorb solar radiation; typically the presence of combustion aerosols increase aerosol absorbing capacity (e.g. VanCuren, 2003)."

"In the principal fire region (see Figure 2), the highest AOT values (0.3-0.6) are ob-

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served from 28 July to 14 August at both the Evora and El Arenosillo sites. During the first part of the period, from 28 July to 4 August, the observed Ångstrom exponent (0.5-0.7) is relatively low, which indicates the prevalence of dust particles. The corresponding single scattering albedo (SSA at 532nm) values range from 0.9 to 0.96 and are consistent with the presence of scattering dust particles."

"As reported by Pace et al. (2006)'s back-trajectory study, the major dust episodes in this region occurred on 16-18 July, 22-24 July and 29 August - 7 September. The predominance of dust particles during this period is also consistent with higher SSA values, from 0.88 to 0.97, found at Lampedusa and Avignon AERONET stations."

"Pace, G., A. di Sarra, D. Meloni, S. Piacentino, and P. Chamard, Aerosol optical properties at Lampedusa (Central Mediterranean). 1. Influence of transport and identification of different aerosol types, Atmos. Chem. Phys., 6, 697-713, 2006."

"VanCuren, R. A., Asian aerosols in North America: Extracting the chemical composition and mass concentration of the Asian continental aerosol plume from long-term aerosol records in the western United States, J. Geophys. Res., 108(D20), 4623, doi: 10.1029/2003JD003459, 2003."

15) Page 4724, Lns 24-27: What are the sources of the anthropogenic pollutants during the stagnant heat wave conditions from 2-13 August, 2003?

Answer: During the heat-wave episode, major sources of anthropogenic pollutants (e.g. aerosols, ozone precursors) are the traffic and industrial activities which are mainly concentrated in the vicinity of large urban and industrial areas. Stagnant meteorological conditions favored the accumulation of pollutants by recycling air masses for several days in an anticyclonic flow over high-emission areas of northern Europe (Vautard et al., 2005).

This has been added in the following sentence: "During this period, aerosol modeling is challenging (Hodzic et al., 2006a) as the aerosol load results from both the accumu-

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lation of anthropogenic pollutants (mainly from industrial activities and mobile sources) during stagnant heat-wave conditions (from 2-13 August) and sporadic wildfire emissions."

16) Pg 4725, Is there any reason for the failure of the model to capture the high AOT in UK and Ireland on the 6th of August 2003 (see Figs. 5 and 6).

Answer: Taking into account wildfire emissions significantly improves model simulations over Northern Europe during the major fire event. Although much higher AOTs are simulated in the presence of the smoke plume, the magnitude of the AOT signal over the UK and Ireland is not fully captured by the model on 5-6 August.

We think that high AOT values observed in satellite data may be contaminated by clouds. In particular, the differences between POLDER and MODIS retrieved AOTs seem to confirm this statement: the majority of the high AOT pixels detected by MODIS are eliminated from the POLDER signal by cloud screening. Also, POLDER visible image for 5 August clearly indicates the presence of several clouds in that region.

This is now explained in the manuscript:

"Also, the AOT retrieval in the presence of clouds is less accurate and contributes to comparison uncertainties. This is most likely the case over the UK and Ireland on 5-6 August, where large discrepancies between model simulated (H_FIRE) and MODIS-observed AOTs can be identified (Figure 5). Differences between POLDER and MODIS AOT retrievals in this region suggest that MODIS data are cloud contaminated. Moreover, the POLDER visible image from 5 August clearly indicates the presence of numerous cloud scatters in this area (see figure 7a of Hodzic et al., 2006a). "

17) Pg 4726, Ln 13-23: This claim does not appear to be quite substantiated by Fig 7.

Answer: We only had this single lidar profile to qualitatively evaluate the simulated aerosol vertical distribution. As this is just a one-case comparison, we made this statement less strong in the paper:

"These results suggest that the considered 'injection height' parameterization gives reasonable simulations of the smoke vertical distribution in the downwind regions."

18) Pg 4729, Section 4.2.3 does not belong here. Since it deals with the effect of biomass burning emissions, it fits better in the aerosol impact section (4.3). Accordingly, you should move it to section 4.3 and possibly denote it 4.3.1. In addition, rather than PM10, why not focus on PM2.5 which is more pertinent to smoke particles- the main focus of this paper. PM10 will include large particles such as dust aerosols.

Answer: Section 4.2.3 deals with changes in aerosol ground concentrations due to biomass burning emissions and is treated just after the effect of wildfires on aerosol total load (AOTs). We find that it is consistent with the content of Section 4.2, which is focused on "Model simulation of smoke emission and transport patterns during the August 2003 heat-wave". Therefore we would like to retain this order of presentation, rather than moving this paragraph into section 4.3 which is reserved to the radiative impact of wildfires.

We agree that PM2.5 is more pertinent to track changes in smoke particles than PM10. We followed the reviewer's suggestion and focused our discussion on PM2.5. Following changes have been made to the manuscript:

"First, directly, the modeled wildfire emissions caused an increase in average PM2.5 ground concentrations from 20 to 200%. The largest enhancement in PM2.5 concentrations stayed, however, confined within a 200 km area around the fire source locations and reached up to 40 ug/m3."

"They resulted in a significant increase in PM2.5 mean concentrations (from 20 to 200%) over several regions in Europe (Figure 9). The largest increase in PM2.5 concentrations is found within a 200 km area around the fire source locations (up to 40 ug/m3), while a more moderate increase (3-5 ug/m3) is observed over the Southern

Mediterranean basin and Benelux countries."

Figure 9 has also been changed accordingly.

" Figure 9: (a) Average PM2.5 ground concentrations (ug/m3) over Europe as simulated by CHIMERE REF run during the first half of August 2003. Increase (%) in PM2.5 concentrations caused by wildfire emissions as predicted by the FIRE (b) and H_FIRE (c) models runs. The relative difference (%) from the reference run is given by ."

19) Pg 4735, section 4.3.2, The atmospheric radiative forcing should be extended to include the fact that the difference in the direct solar radiation between the top of the atmosphere and the surface was done for clear sky (with and without biomass burning aerosols).

Answer: It is now included in section 4.3.2 : "In this study, the daily averaged radiative forcing of fire emitted particles is calculated in clear sky conditions by integrating over time and wavelength intervals the spectral irradiances simulated with and without fire emitted aerosols."

20) Pg 4736, Ln 3, replace "reinforced" by "reinforce" Done.

21) Pg 4746, Place an asterisk beside AOT in the title of the Table to imply that there is a footnote below the Table. Done.

2. SPECIFIC ANSWER TO REVIEWER #2:

Answer to Reviewer's General Comment:

As suggested by both reviewers the objectives and new work performed in the present study have been pointed out more clearly in the manuscript. Specifically, the following paragraph has been added:

"Hodzic et al. (2006a) reported large inconsistencies in predicted aerosol concentrations and optical properties over Europe during summer 2003 caused by unaccounted emissions from wildfires. The present study extends this work by developing a new

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modeling framework that includes wildfire emissions and their effect on air quality. The objective of the paper is twofold: (i) First, a simple parameterization of smoke emission and transport is presented and evaluated with observations; (ii) Second, the effects of smoke emissions on air quality in Europe are examined during the summer 2003 fire season, including both the direct impact on ground concentrations of pollutants and the indirect impact on photolysis rates and atmospheric radiative forcing."

Minor Comments:

1)p. 4713, l. 3-8: Fig. 1 should be referenced here. Done.

2)p. 4714, l. 17: change "can be overestimated" to "can be an overestimate". Done.

3)p. 4726, l. 16-17: the authors should consider directly reproducing Fig. 6 from Hodzic et al. (2006a). Answer: Direct comparison of model results with lidar data is not possible as the arbitrary units are used to indicate the relative strength of the lidar backscatter. Also, the Figure 7 of the present paper is more suitable in this study as it inter-compares 3 model simulations which cannot be displayed in Figure 6.

4)Section 4.3.1: The authors demonstrate the effect wildfire particulate matter on photolysis rates by comparing the REF and H_FIRE runs. Furthermore, CHIMERE reference model ozone concentrations are compared with measurements. Is this reference model different from REF? Is online ozone chemistry also included in the REF and H_FIRE model runs? If yes, ozone concentrations from both runs should be compared in order to directly identify the wildfire effect on ozone.

Answer: The aerosol-induced changes in photolysis rate coefficients were calculated offline, and therefore do not give direct estimate of changes in ozone production. Model improvements are underway to fully couple the aerosol radiative effects into the chemistry and transport model.

5)pp. 4736, l. 2-4: revise verb "can reinforced" in this sentence. Answer: The sentence has been changed to: "This heating could have contributed to reinforcing the atmo-

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spheric stability and to maintaining the heat-wave conditions during summer 2003."

6)For many of the figures, fonts used for labels and numbering are too small even if the figures are scaled to ACP format (e.g., Figs. 4, 5, 6, 11, 12, 14). I would suggest using only one larger color bar for all model subfigures in Fig. 6 and, similarly, one for MODIS and one for the model results in Fig. 5.

Answer: Fonts have been made bigger in figures 4, 11, 12, 14. We could not produce a better plot for figures 5 and 6, and we kept the same presentation as in our previous paper (Hodzic et al., 2006). The Figure 13 has also been updated due to an error that occurred in the unit scale.

Additional minor changes (style and typography errors) were also performed in the new manuscript:

Abstract:

"The model has been improved to take into account a MODIS-derived daily smoke emission inventory as well as the injection altitude of smoke particles."

"The simulated aerosol optical properties are put into a radiative transfer model to estimate (off-line) the effects of smoke particles on photolysis rates and atmospheric radiative forcing. We have found that the simulated wildfires generated comparable amounts of primary aerosol pollutants (130 kTons of PM2.5, fine particles) to anthropogenic sources during August 2003, and caused significant changes in aerosol optical properties not only close to the fire source regions, but also over a large part of Europe as a result of the long-range transport of the smoke."

Section 1.1: Challenge of modelling fires: parameterizations and uncertainties

"To accurately simulate smoke transport in the atmosphere and to quantify its radiative impact in meso-scale models, the fire emissions and their transport patterns need to be accurately estimated."

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Section 2.1 : Fire emission inventory

“In this study, daily fire emissions of particulate matter and their locations during summer 2003 in Europe were estimated using a methodology similar to the one described by Wiedinmyer et al. (2006). Briefly, the information required to estimate emissions from fires includes (i) the location and date of the fire event, (ii) the area burned, (iii) the fuel loading factors (mass of biomass per area), (iv) the combustion efficiency, or the fraction of biomass fuel burned, and (v) the emission factors (mass of species emitted per mass of biomass burned). For this study, these parameters have been determined by combining data available from several satellite products.”

“The land cover type at each fire location was identified by the MODIS Land Cover Type product (http://edcdaac.usgs.gov/modis/mod12q1v4.asp). The identified IGBP land cover classifications are further assigned to 5 generic land cover types (boreal forest, temperate forest, tropical forest, woody savanna, grasslands and savanna, and croplands).”

“However, comparisons with other available emissions inventories (i.e., van der Werf et al., 2006) show that these estimates are within 50% of other estimates for most emitted species.”

“The daily estimated fire emissions are produced with 1 km spatial resolution. For the purposes of this study, the emissions have been integrated over the CHIMERE regional 0.5°x0.5° longitude-latitude grid” Section 3.4: Experiment design:

“The first simulation (referred to as the REF run) does not account for smoke emissions and is considered the reference case since it has been studied extensively and evaluated in a previous paper (Hodzic et al., 2006a).”

Conclusion:

“This impact was significant not only in magnitude, but also in terms of its spatial (continental) and temporal (seasonal) extent.”

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“The radiative effects of aerosol concentrations and their controls on ozone formation should also be included in future ozone reduction scenario calculations as the particulate matter controls are expected to increase surface ozone concentrations over polluted regions in Europe.”

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 4705, 2007.