Anonymous Referee #1

The manuscript presents a modelling study of the photochemical impact of a major wildfire event in Russia during summer of 2006. The subject of the paper is certainly within the scope of ACP and specifically addresses the question on how the emissions from wildfires and from biogenic sources interact and affect ozone production rates. Before considering publication on ACP, I suggest the authors to consider the following points:

We would like to thank Referee #1 for his very constructive comments and recommendations. The proposed by him new literature, published after the first submission of this paper, proved very useful. We have corrected and improved the paper by incorporating most of the reviewers' comments, in the revised version. However, it was not feasible to consider all the issues mentioned in the review paper of Jaffe and Wigder (2012) that could be a research area in a future study.

We would like to underline that this case study refers to spring and not summer fires of 2006. According to Sofiev et al. (2006), there is a fundamental difference between spring and summer fires of 2006. During the spring widespread wildland fires over Western Russia, the burning material mainly consisted of previous-year grass remnants, which were dried up when the fires begun. In particular, the daily distribution of the total Fire Radiative Power (FRP) from each land-use class revealed that the case of spring fires 2006 represents mainly a dry grass fire. However, emissions from forest and mixed areas also exist. On the contrary, summer fires 2006, took place in Northern Europe and South-Eastern Europe. The burning material was a mixture of grass, agricultural remnants (mainly in the south), but also bog and forest trees (mainly in the north). Given that the burning material determines the magnitude and the composition of fire emissions thus, photochemistry, our results must be interpreted based on the choice of this specific case study.

• I believe the title is not accurate. The manuscript reports on sensitivity tests perturbing fire and biogenic volatile organic compounds (VOC) sources, thus the word "anthropogenic" sounds a little bit out of context at the end of the reading. I suggest modification of the title with "Ozone production from the interaction of wildfire and biogenic emissions: a case study in Russia during summer of 2006"

The title was corrected accordingly to: "Ozone production from the interaction of wildfire and biogenic emissions: a case study in Russia during spring of 2006"

• Introduction. I suggest adding this recent reference, which is a review of the subject of the paper: "Jaffe and Wigder (2012), Ozone production from wildfires: A critical review, Atmospheric Environment 51, pp. 1-10." This other paper addressed the issue of the interaction of wildfire emissions with BVOC

and urban areas with respect to the production of ozone: "Junquera et al., Wildfires in eastern Texas in August and September 2000: Emissions, aircraft measurements, and impact on photochemistry, Atmospheric Environment 39 (2005) 4983–4996." It could/should be used as a term of comparison with results presented here. E.g. the authors report a major contribution of isoprene BVOC oxidation in the fire plume of 63% and 33% over a forest and near urban area, respectively. Moreover, I believe there are also other studies following the Russia major wildfire event of summer of 2010. E.g., from a quick search on ACP: "Atmospheric impacts of the 2010 Russian wildfires: integrating modelling and measurements of an extreme air pollution episode in the Moscow region, I. B. Konovalov, M. Beekmann, I. N. Kuznetsova, A. Yurova, and A. M. Zvyagintsev, Atmos. Chem. Phys., 11, 10031-10056, 2011.

The proposed references (Konovalov et al., 2011; Jaffe and Wigder, 2012; Junquera et al., 2005) are now included in the Introduction. Also, some other recent references have been added (Alvarado et al., 2010; Singh et al., 2010; Alvarado and Prinn, 2009; Nassar et al., 2009; Ø. Hodnebrog et al., 2012). However, the comparison with the results of the proposed papers is not straightforward since the studies of Jaffe and Wigder (2012) and references therein concentrate mainly on fires in boreal and tropical forests while the 2010 Russian wildfires (Konovalov et al., 2011) are characterized by increased peat fires emissions and massive large-scale burns under anomalously high temperatures for the season and very dry conditions. The case we considered was mainly created by numerous small-scale dry-grass fires, largely man-made, that occurred under quite typical latespring conditions (Sofiev et al., 2006). In that light, our findings rather complement the above studies in terms of prevailed conditions and types of fires. Regarding the comparison with the findings of Junquera et al. (2005), this is also not straightforward since the contribution of isoprene's oxidation in O<sub>3</sub> formation in fire plumes (near forest or urban area) has been estimated in a different environment (US). Nevertheless, the application of the Factor Separation (FS) analysis gave us the opportunity to appoint in the revised manuscript the interaction effect of fires and biogenic emissions over Western Russia and Eastern Europe.

Methods. In section 3.2 and 3.3 the impact of wildfires and BVOC emissions is studied. The method consist in using differences among these simulations: A. Reference: all emissions included B. NoFIRES: wildfires emissions off C. NoBIOG: BVOC emissions off D. NoFIRES+NoBIOG: wildfires and BVOC emissions off The impact of wildfires is assessed using the difference of runs A-D, that of BVOC using the difference A-C. Simulation D is discussed by the end of section 3.3, with no clear target. In my opinion, this method is not correct. According to the Factor Separation analysis framework presented by Stein and Alpert ("Factor Separation in Numerical Simulation", J. Atmos. Sciences, Vol. 50, No. 14, 1993), when the effect of two interacting factors are examined (as in this case), the following differences of runs listed above should be used: B – D for effect of wildfires alone C – D for effect of BVOC alone A – (B+C) + D for the combined effect of wildfires and BVOC The difference between A and C or D is useful when only ONE factor is under investigation. However, as the

authors state from the title, the objective of this paper is to study the interaction of wildfires and BVOC emissions, thus the Stein and Alpert (1993) framework should be applied. I believe all results presented in sections 3.2 and 3.3 should be reformatted in this context.

The method of Stein and Alpert (1993) has been applied and the results of section (3) have been reorganized. In the revised manuscript, section 3.1 discusses the episode analysis, section 3.2 the evaluation of the  $O_3$  predictions, section 3.3 discusses the results of the FS analysis and finally section 3.4 the results of the sensitivity tests. The presentation of the FS method and the necessary simulations are presented in section 2.

In our study, the interaction of two factors, biogenic and fire emissions, is investigated. Following the FS technique, four simulations have been performed (Table 1). The simulations results are denoted by f while the variables f' denote the pure contributions from fires  $(f'_F)$  and biogenic  $(f'_B)$  source categories. At this point we would like to mention that  $f'_F$  (pure contribution of fire emissions) is estimated as C-D and not B-D. Similarly, the  $f'_B$  (pure contribution of biogenic emissions) as B-D and not C-D. The variable  $f'_{BF}$  represents the contributions due to the interaction of the two sources while  $f'_0$  is the O<sub>3</sub> unrelated to either biogenic or fire emissions. The total or "actual" contribution of the two source categories is also examined in the revised manuscript as discussed in Tao et al. (2005). For a source category, this is defined as the difference between the simulation results considering all source categories  $(f_{BF})$  and those with all but this source considered  $(f_B \text{ or } f_F)$ . The anthropogenic emissions are included in every simulation as background.

In the first submitted paper, all the runs presented in Table 1 have been performed but the discussion was mainly focused on the total (actual) contribution of fires and biogenic emissions. Below the revised section 3.3 is presented.

Simulation	Simulation	Emissions	
	results		
Reference (A)	$f_{BF}$	Anthropogenic, biogenic, fires	
NOFIRES (B)	$f_B$	Anthropogenic, biogenic	
NOBIOG (C)	$f_F$	Anthropogenic, fires	
NOFIRES+NOBIOG	$f_0$	Anthropogenic	
(D)			
Contribution of source categories			
$f_{BF}-f_F$		Total contribution of biogenic emissions	
$f_{BF}$ - $f_B$		Total contribution of fires emissions	
$f_0 = f_0$		O <sub>3</sub> unrelated to either biogenic or fire emissions	

**Table 1:** Summary of model runs performed for the application of the FS technique and definitions of the various contribution components

$f_B' = f_B - f_0$	Pure contribution of biogenic emissions	
$f_{\scriptscriptstyle F}^{'}=f_{\scriptscriptstyle F}-f_{\scriptscriptstyle 0}$	Pure contribution of fire emissions	
$f'_{BF} = f_{BF} - (f_B + f_F) + f_0$	Synergistic effect due to fires and biogenic	
	emissions	

## 3.3 FS technique - Results and discussion

In order to calculate the pure, synergistic and total contributions of biomass burning and biogenic emissions in maximum surface  $O_3$ , the results of the NOBIOG, NOFIRES and NOFIRES+NOBIOG simulations are used, at that time the maximum hourly surface  $O_3$  concentration appears in each grid cell in the reference run.

### 3.3.1 Pure contribution from individual source category

In the framework of this study, the pure contribution represents the full potential of each source category (biomass burning or biogenic emissions) to produce  $O_3$  in the presence of anthropogenic sources. In Figure 10, the pure contribution of fire emissions on maximum hourly surface  $O_3$  concentrations is presented for each day of the period 2-7 May. It is evident that the pure contribution of fire emissions determines the spatial distribution of surface  $O_3$  over large parts of Europe (pp. 3499, Fig. 5). According to the analysis presented in section 3.1, fire emissions are strong  $NO_X$  emitters and thus provoke  $O_3$  titration near the source. Close to the burnt areas and in areas characterized by low VOC/NO<sub>X</sub> ratios, the negative contribution reaches up to 35 ppb. At the same time fires are positive contributors downwind, in more aged plumes, enhancing  $O_3$  production. The maximum contributions occur in areas where the environmental conditions are ideal for  $O_3$  production. Specifically, over Western Russia, Eastern and Northern Europe, the fires' pure contribution is up to 40-45 ppb. The higher potential of  $O_3$  production towards the end of the episode is related with the decline of the fire intensity and the elimination of  $NO_X$  emissions in the VOC-sensitive environment.

The pure contribution of biogenic emissions is presented in Figure 11. In the presence of anthropogenic emissions, the expected  $NO_X$ -limited environment over Scandinavia, Eastern Europe and Western Russia determines the role of biogenic emissions. The pure contribution of biogenic emissions over these areas is negligible or even negative (-1 ppb). The areas characterized by negative contributions coincide with the areas of enhanced biogenic activity (pp. 3495, Fig. 1). For the NOFIRES simulation and during the days of maximum photochemical activity, isoprene contributes to the VOC-OH oxidation rates (not shown), up to 25% over Western Russia and 10% over Eastern Europe. These fractions are higher compared to the reference case, as the presence of fire VOC emissions depresses OH levels, thereby reduces the reactivity of BVOC emissions. However, under low  $NO_X$  concentrations (NOFIRES simulation), the peroxy radicals, produced by VOC-OH oxidation, are removed from the system through radical-radical reactions, rather than participating in chain reactions converting NO to  $NO_2$ . Only over more populated areas (e.g. Moscow), the pure contribution is positive, of the order of 1 ppb.

Over the rest of Europe, the pure contribution of biogenic emissions does not exceed 2-3 ppb. The positive increase near UK, on 3 and 4 May, is attributed mainly to the interaction of BVOC emissions and ports activity. Despite the fact that over the southern Europe, biogenic activity, exhibit the highest values in Europe, the lack of essential NOx for efficient  $O_3$  production leads in relatively small  $O_3$  increases. The same result (e.g. for Spain) has been also discussed by Curci et al. (2009). The negative contribution over Morocco and Tunisia is associated with ozonolysis of isoprene and terpenes and mainly reflects the poor model representation of anthropogenic emissions in these areas.

#### 3.3.2 Synergistic effect between biogenic and biomass burning emissions

The synergy between a source and other source categories indicates if the mixing of both categories tends to limit or enhance the potential of any single source category in producing  $O_3$ . In this study, the synergistic effect between biogenic and biomass burning is presented in Figure 12. During the days of intense fire activity coinciding with low biogenic activity (up to 2 May), the synergistic effect is small, about 1 ppb, or even negligible. As the fires  $NO_X$  plumes disperse over regions which also emit biogenic emissions (Western Russia and Scandinavia), they provide the necessary fuel for the peroxy radicals, produced by BVOC-OH oxidation, to participate in chain reactions converting NO to  $NO_2$ . The negative synergy effect over more populated areas (Moscow) is attributed to the addition of fire  $NO_X$  emissions in an already urban VOC-environment and suppresses further  $O_3$  production by 1 ppb.

The synergistic effect becomes more apparent after 3 May over Western Russia and Eastern Europe, under the favorable conditions for the VOC chemistry discussed in the previous section, and it is maximized between 4 and 6 May. This effect becomes apparent in areas designated by enhanced biogenic activity but also by ideal environmental conditions to produce  $O_3$ . For example, the synergy between the two source categories over northern Europe on 6 May is attributed to the enhanced biogenic activity over this area although the pure effect of biogenic emissions is negative or negligible (Fig. 11). As the fire plume passes over northern Europe is enriched by reactive BVOC emissions and provokes a further  $O_3$  increase by up to 6 ppb. But also downwind of the fire plumes, in areas characterized by favorable meteorological conditions and higher VOC/NO<sub>X</sub> ratios (> 5 ppbC/ppb), the interaction of BVOC and fire emissions enhance the potential of the BVOC in  $O_3$  formation by 8 ppb. On the contrary, during 8 May (not shown), even though biogenic emissions over Western Russia are comparable with the emissions during the previous days (2.5-3 mg m<sup>-2</sup> day<sup>-1</sup>, not shown), their synergistic effect on surface  $O_3$  does not exceed 1 ppb mainly due to low fires activity (Fig. 3) and low  $NO_X$ concentrations.



*Figure 10*: Pure contribution of fire emissions on maximum hourly surface  $O_3$  (ppb) during the period 02/05-07/05/2006.



*Figure 11*: Pure contribution of biogenic emissions on maximum hourly surface  $O_3$  (ppb) during the period 02/05-07/05/2006.



*Figure 12*: Synergistic effect of biogenic and fire emissions on maximum hourly surface  $O_3$  (ppb) during the period 02/05-07/05/2006.

#### 3.3.3 Total contribution from individual source category

The total (actual) contribution of fire emissions to maximum surface  $O_3$  under the presence of both anthropogenic and biogenic source categories (Figure 13) does not significantly differ from the pure one (Figure 10) suggesting the critical role of biomass burning emissions in the examined area. Small increases of the order of 5 ppb are estimated in areas where the synergistic effect of the two emission categories is positive.

Regarding biogenic emissions, their total contribution in  $O_3$  production (Figure 14), was found to almost coincide with the synergetic effect between the two sources and thus, it is completely different compared to the pure one. Therefore, for this source category, the presence of fires is critical for determining its potential in producing  $O_3$ . In particular, in areas where the pure contribution of biogenic emissions was found negative, the synergetic effect of these two emission categories reversed their final impact on the  $O_3$ production. During the days of maximum photochemical activity, the positive total contributions are emphasized in the rich-in  $O_3$  plumes and reaches up to 8 ppb.

Isoprene was found to be the major contributor to the  $O_3$  increases, by 80% (up to 6 ppb). Terpenes and BOVOC emissions have lower contribution, 0.5-1 ppb, likely because of lower emissions for the terpenes case and the low reaction rates for the BOVOC category. Even though the emission load of isoprene and BOVOC categories is comparable, the methanol's (99% of BOVOC) low oxidation rate by OH radical (1,600 ppm min<sup>-1</sup> compared to isoprene's 147,600 ppm min<sup>-1</sup>) results in low  $O_3$  productivity. Due to this low reactivity,  $O_3$  increases are evident only over areas with maximum BOVOC emissions (> 0.5 mg m<sup>-2</sup> day<sup>-1</sup>, Fig. 11d). On the contrary, the most reactive compounds, isoprene and terpenes, interact substantially with fire emissions and exhibit additional areas of influence.



*Figure 13*: Total contribution of fire emissions on maximum hourly surface  $O_3$  (ppb) during the period 02/05-07/05/2006.



*Figure 14*: Total contribution of biogenic emissions on maximum hourly surface  $O_3$  (ppb) during the period 02/05-07/05/2006

• The authors should mention somewhere in the paper that they neglected the effect of concomitant aerosol emissions on photolysis rates. As also reported in the review of Jaffe and Wigder (2012), the "obscouring" effect of aerosol may decrease the ozone production at the surface by up to 20%.

The text has been revised accordingly:

"..... for each day of the simulation period. The aerosol impact on photolysis rates has not been taken into account although the "obscouring" effect of aerosols (Tzanis et al., 2009; Konovalov et al., 2011) may decrease the ozone production at the surface by up to 20%" (Jaffe and Wigder, 2012).

• Mixing vs. photochemistry. A good discussion point might be an analysis of the relative attribution of ozone production rates along the plume path of mixing with background airmasses and photochemical processing inside the plume of original emissions. This is an interesting point raised in the Jaffe and Wigder (2012) review, which is still unclear in the existing literature and thus might be an innovative contribution of this paper.

The application of the Factor Separation (FS) analysis in the revised manuscript gave us the opportunity to appoint the interaction effect of the mixing of fires and biogenic emissions. The analysis of FS method revealed that the mixing of the gases of fire plumes with biogenic emissions tends to enhance the O<sub>3</sub> forming potential of each single source category by up to 8 ppb in the area. However, the peak (spatially) synergistic effect does not necessarily coincide with areas characterized by peak biogenic emissions strength. It also depends on the environmental conditions that favor O<sub>3</sub> increase namely the meteorological conditions and the O<sub>3</sub>-VOC-NOX sensitivity of the areas crossing the fire plumes. Over the fires dominated area, which is not so densely populated, a significant fraction of the VOC-OH oxidation rate 30-40% is due to the abundance of the CO fire emissions. Only a small fraction 5% is attributed to isoprene. Downwind of the fire plume, the less reactive CO emissions, compared to VOC, arrive intact, and contribute to the VOC-OH oxidation rates by 50%. During the days of maximum photochemical activity coinciding with days of maximum biogenic activity, isoprene contributes to the VOC-OH oxidation rates, up to 15% over Western Russia and 5% over Eastern Europe.

In this study, the fires mostly evolve in a region with relatively low anthropogenic emission load. The effect of anthropogenic emissions along the fire plumes as well as the relative attribution of ozone production rates along the plume path is indeed an interesting issue to consider in a future study.

• Role of PAN. Maybe a more detailed analysis of the evolution of PAN concentration is needed. Indeed, PAN is believed to be the main reservoir species regenerating NOx in the plume even weeks after the injection and is thus a key driver of the evolution of the VOC/NOx ratios in the plume as it travels. PAN is currently only mentioned in the sensitivity test on NOx/CO ratios in

emissions. In the case of low NOx/CO emission ratio in fires the authors calculate that PAN makes a fraction of NOy of 25%, while in the high NOx/CO ratio the share is 40%. What is the magnitude in ppb of PAN in the two cases? Indeed, from literature (see Jaffe and Wigder, 2012, and references therein) it seems like PAN should be much higher in the low NOx/CO ratio case, because of the enhanced abundance of oxygenated compounds. Please clarify this point.

After the reviewer's suggestions, we included in the analysis of the reference run (NO<sub>X</sub>/CO=0.06) (section 3.1) results of PAN predictions and a relevant discussion. We also enriched the relevant results presented for the case of low NO<sub>X</sub>/CO ratio (0.025) in the section 3.4. Also, a short discussion regarding the PAN decomposition and the potential contribution to the O<sub>3</sub> increase along the plume path was added. Regarding the fraction of 40% in the high NO<sub>X</sub>/CO ratio, in the first submitted paper, the reviewer is correct, this was a mistake that has been corrected. We would also like to mention that in the revised manuscript NOy is considered as the sum of NO<sub>X</sub>+PAN+HNO<sub>3</sub>+nitrates, while, in the first manuscript NOy neglected NO<sub>X</sub>.

According to the revised manuscript, in the case of high NO<sub>X</sub>/CO ratio (0.06), the model predicts PAN concentrations up to 3 ppb in the area dominated by fire emissions. Close to the fires and in fresh plumes, the PAN/NOy and HNO<sub>3</sub>/NOy fractions are up to 5% and 25%, respectively while the greater fraction is for NO<sub>X</sub> (80%). In the case of low NO<sub>X</sub>/CO molar ratio (0.025), the predicted PAN concentrations are higher, up to 15-20 ppb, despite the fact that agricultural residues (NO<sub>X</sub>/CO=0.025) emit lower NO<sub>X</sub> emissions than grass (NO<sub>X</sub>/CO=0.06). In this case, the PAN/NOy and HNO<sub>3</sub>/NOy fractions are up to 50-60% and 20-30%, respectively.

• On the NOx/CO ratio. In the reference case a high NOx/CO ratio of 0.06 is chosen. Actually, the region of interest may be considered more similar to a boreal environment, where usually lowest values of combustion efficiencies are reported (see again references in Jaffe and Wigder, 2012), which are associated with lowest NOx/CO ratios, with respect to other environments (e.g. tropical, savannah, etc. Shouldn't be the low NOx/CO ratio emission of 0.025 be used as the reference in this case?

We would like to repeat that the case study we considered refers to spring and not to summer fires of 2006. According to Sofiev et al. (2006), there is a fundamental difference between spring and summer fires 2006. During the spring widespread wildland fires over western Russia, the daily distribution of the total FRP release from each land-use class (forest, grass, mixed) revealed that the case of spring fires 2006 represents mainly a dry grass fire (Sofiev et al., 2009) and not boreal fires (Fig. 2 *in Sofiev et al., 2009*). For that reason we have selected the high NOx/CO ratio, corresponding to grass, for the reference case. The lower NOx/CO emission ratio corresponding to agricultural residues is used to reveal the sensitivity of  $O_3$  simulations on the emission properties.

• Sensitivity tests. This tests may be quite useful at least for two reasons: (1) estimate uncertainty on the ozone production assessment present in the result section, and (2) identify the single parameter, if any, which dominates the simulation uncertainty. The authors describe the results of tests without showing any figure/table and do not draw any clear conclusion related to the two points just mentioned. I suggest showing some results (maybe in the supplementary online material, to avoid an excessive number of figures in the main paper) and better clarifying what the reader may learn from these tests. The latter point should be concisely repeated in the conclusions and in the abstract, because it could be a good contribution of the paper to the scientific community.

We agree with the reviewer, therefore, in the revised manuscript, the discussion of the sensitivity studies in relation to the reference run has been reorganized. In particular, we have added Table 3 in section 3.4, presenting the maximum positive and negative  $O_3$  differences between the sensitivity and the reference runs following the methodology of R. von Kuhlmann et al. (2004, in ACP).

It is shown that the maximum  $O_3$  predictions are very sensitive to emission characteristics and specifically to the molar  $NO_X/CO$  ratio. The largest positive differences occur when a lower NO<sub>X</sub>/CO molar ratio is applied (AGRIC run considering agricultural residues as burning material, NO<sub>X</sub>/CO=0.025) and equal 107.8  $\pm$  53.8 ppb. This result implies that the chemical composition of fire emissions strongly influence the O<sub>3</sub> formation in a certain environment. The higher increases are predicted during the days of high fire intensity and over the fires dominated areas. The cut-off of fire emissions by 20% (EMISS0.8) ends in maximum  $O_3$  increases of  $7.8 \pm 3.3$  ppb in relation to the reference run. These increases take place close to the fire spots where O<sub>3</sub> titration by NO prevails. Negative differences of  $-5.4 \pm 1.9$  ppb are predicted inside the more aged plumes (Eastern and Northern Europe, Western Russia) whose chemistry is primarily controlled by NO<sub>x</sub>availability. For the F1 scenario (the daily maximum injection height is determined by the calculated daily maximum PBL height over the burnt area), the maximum positive difference on hourly surface  $O_3$  concentrations is  $4.2 \pm 1.8$  ppb and appears in the vicinity of the fires. The release of fire emissions up to higher altitudes favors the O<sub>3</sub> enhancement within the PBL which under favorable conditions is mixed downward increasing surface concentrations. As in the previous case but lower in magnitude, negative differences of  $-1.8 \pm 0.7$  ppb are predicted inside the more aged and rich-in O<sub>3</sub> plumes. The differences are more pronounced for the F2 scenario (assesses the impact of possible crown fires; 60% of the emissions are injected above the top of the PBL up to 5 km and the rest 40% represents emissions from surface fuels and are uniformly distributed up to a representative average PBL height). The lower NO<sub>X</sub> emissions released at surface and inside the PBL, in relation to the reference run, enhance O<sub>3</sub> production in fires dominated areas by  $24.7 \pm 3.9$  ppb. Negative differences of  $-6.5 \pm 3.5$ ppb are estimated in NO<sub>X</sub> sensitive areas (rich-in O<sub>3</sub> plume along the SSE-NNW axis).

Parts of the above discussion will be included in the abstract and the conclusions. According to reviewer's suggestions, we also provide Figs. S1-S4 presenting the  $O_3$  concentration differences between the sensitivity runs and the reference run at the time the maximum hourly surface  $O_3$  concentration appears in the reference run. These figures could be shown as supplementary online material.

**Table 3:** Maximum positive and negative O<sub>3</sub>differences between the sensitivity runs and the reference run.

	Positive differences (ppb)	Negative differences (ppb)
AGRIC	$107.8 \pm 53.8$	$-6.8 \pm 3.4$
EMISS0.8	$7.8 \pm 3.3$	$-5.4 \pm 1.9$
F1	$4.2 \pm 1.8$	$-1.8 \pm 0.72$
F2	$24.7 \pm 3.9$	$-6.5 \pm 3.5$



Fig. S1: O<sub>3</sub> concentration differences between the AGRIC sensitivity run and the reference run at the time the maximum hourly surface O<sub>3</sub> concentration appears in each grid cell in the reference run

# **Results of sensitivity runs**

02/05/2006

03/05/2006



Fig. S2: O<sub>3</sub> concentration differences between the EMISS0.8 sensitivity run and the reference run at the time the maximum hourly surface O<sub>3</sub> concentration appears in each grid cell in the reference run



Fig. S3: O<sub>3</sub> concentration differences between the F1 sensitivity run and the reference run at the time the maximum hourly surface O<sub>3</sub> concentration appears in each grid cell in the reference run



03/05/2006



Fig. S4: O<sub>3</sub> concentration differences between the F2 sensitivity run and the reference run at the time the maximum hourly surface O<sub>3</sub> concentration appears in each grid cell in the reference run