Atmos. Chem. Phys. Discuss., 11, C12503–C12518, 2011
www.atmos-chem-phys-discuss.net/11/C12503/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evolution of trace gases and particles emitted by a chaparral fire in California" *by* S. K. Akagi et al.

S. K. Akagi et al.

bob.yokelson@umontana.edu

Received and published: 30 November 2011

Reply to Referees #1, #2 and #3. We thank all Referees for their helpful and thoughtful comments. Our response to each specific point made by the Referees follows:

Anonymous Referee #1 R1.1. Review of "Evolution of trace gases and particles emitted by a chaparral fire in California" by S.K. Akagi, et al. submitted for publication in Atmospheric Chemistry and Physics.

The authors present the results of new in-situ field measurements of several ambient gases and particle properties inside a smoke plume from a prescribed fire of chaparral fuels in California in November 2009. Measurements made directly at the fire source were used to characterize the primary emissions and determine the emission

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

ratios (ERs) and calculate emission factors (EFs) of 18 gas-phase species as well as particle composition and optical properties. The uniquely isolated fire plume was then systematically measured at discrete distances downwind of the source in order to study the gas-phase photochemistry and particle transformations within the "aging" fire plume. The collective measurements clearly indicate that OH oxidation within the plume resulted in a decrease in the primary reactants (e.g., C2H4 and NOx) and the subsequent photochemical formation of secondary products (e.g., O3, HCOOH, CH3COOH and PAN) on the timescale of 0-4.5 hours after initial emission at the fire source. Notable results of the particle measurements include the overall decrease in OA with plume aging while particle scattering (bscat) increased. The authors compare the chemical evolution of the Williams Fire to that of a previously documented tropical biomass burning plume(s) in Mexico in March 2006. The measurement techniques and scientific methods included in this analysis are robust and the data collected represents a unique and valuable dataset. The findings of this analysis are of significance to a wide audience, particularly to those interested in the composition, chemistry and climate impacts of biomass burning. The topic is certainly relevant to readers of ACP. I recommend publication after revision.

R1.2. General Comments:

1.2.1. It would be incredibly useful if all of the findings discussed in sections 3.2.1 to 3.4 were summarized in a new table(s). This table should include the fire-average initial molar emission ratios and standard deviations included in Figures 5-12 and the final measured or fitted NEMRs at age = 4-4.5 hours for all the relevant species/parameters. Additional columns could include the net and relative changes of the Williams Fire. Since the Yokelson (2009) analysis of the Mexican plume is repeatedly compared to the Williams Fire, I suggest adding a column to the new table that has the relative change of the Yucatan plume at t = $\sim$1.5 hour for ease of comparison. This would also simplify and reduce the amount of repetitious text throughout Sect. 3.2 to 3.4.2.

Authors Response. This is a great suggestion that would help emphasize changes in

ACPD

11, C12503–C12518, 2011

NEMR observed in aged smoke. We will work on implementing a new, combined table displaying the content recommended by Referee #1.

1.2.2. The data presented in Table 2 isn't directly applicable to the subsequent discussion. The slope-based source ERs and uncertainty in Table 2 are not used in the discussion of the plume evolution, the mean and standard deviations are. Furthermore, Burling (2011) has already published MCE and EFs for the Williams Fire given in Table 2. Since there is little to no discussion of the MCE and EFs in the text, there is little need for them to be included here. I suggest removing Table 2, especially if the new table mentioned above is included.

Authors Response. The slope-based source ERs are the best estimate of the fire-average ER and an important result that may provide useful context for the reader to see how our chaparral fire ERs compare with others in the literature (computed in the same way). Additionally, we want to emphasize that our source ERs for the Williams Fire are indeed quite typical of chaparral fuel types. These slope-based ERs also serve as the "starting point" used to measure downwind changes, not the mean ER. The standard deviation in the mean of the ten source samples, however, is used as the initial uncertainty in the starting point for the individual downwind samples and is shown at an estimated time since emission of 0 h in all aging plots. Referee #1 makes a good point in that the error calculated as the uncertainty in the slope is minimally discussed in the body of this work, and, also appears in the work of Burling et al. (2011). This uncertainty is most appropriate for comparing between fires. Thus, we propose eliminating Table 2 and limiting our discussion to the representativness of the Williams Fire as a typical chaparral fire, while referring the reader to Burling et al. (2011) for a more detailed comparison of these ER and EF to other fires. In Section 3.1 we plan to introduce a new Table 2 as suggested in Referee #1 Comment 1.2.1. The new table will once again show the slope-based ER, along with the standard deviation in the mean of the ten source measurements.

1.2.3. The purpose of Sect 3.1, other than an introduction to Table 2, is unclear. The

ACPD

11, C12503–C12518, 2011

authors discuss the difference in the uncertainties in ERs using the slope-based approach versus the standard deviation of the mean; however, it isn't entirely clear why one uncertainty is a better metric than the other based on the amount of plume mixing. I understand that it is difficult to estimate mixing, but what (if anything) can be learned from the long-lived species with relatively large emissions in the fire such as CH4 and C2H2? Wouldn't a decrease in these long-lived species inside the plume point to mixing of the fire plume with background air (i.e., dilution) since chemical destruction/production on the <4 hour timescale be negligible?

Authors Response. To reduce confusion and to minimize potentially distracting tangential topics, we have removed the discussion on plume mixing and focus this section on the initial emissions from the Williams Fire and how these emissions are characteristic of a typical chaparral fire. All discussion of downwind NEMR and using the slope-based ER as a "starting point" for downwind analysis has been moved to Section 3.2. In Section 3.2 we will also describe our use of the standard deviations of the mean (of the ten source measurements) to initialize the plume evolution plots.

4. I would suggest using the more specific "elapsed time" or "time since emission" rather than the general term "age," which has a similar connotation as photochemical age and atmospheric lifetime, etc.

Authors Response. We will incorporate "time since emission (h)" as a straightforward term describing the amount of time that a given sample of smoke has been present in the atmosphere.

5. It can get confusing for the reader when the absolute ERs and NEMRs and the relative changes with aging are discussed in the text in percents, fractions and factors. For example, on P22503 the discussion of the change in organic acid ratios are given in Fig. 6 and Table 2 as a straight fraction but referred to in the text as a percent. The net changes in the organic acids are given as factors while the net ΔHCHO/ΔCO is given as a percent. I suggest limiting the use of percents except where necessary.

ACPD

11, C12503–C12518, 2011

Authors Response. We agree that we should try to maintain some consistency when reporting relative changes in ERs and NEMRs. We will minimize the usage of percents throughout the text and report all relative changes as fractions and factors.

6. The role of HONO is not discussed in this manuscript even though it had an appreciable EF. HONO would be an additional source of OH in the daytime fire plume. The authors state that the difference in estimated OH between the Williams Fire and the African+Mexican fires is due primarily to the amount of solar radiation, but how comparable were the sources of HONO?

Authors Response. The ER for ΔHONO/ΔCO was actually lower in the Yucatan (2.8E-3) and Africa (1.22E-3) than in this work ($\sim$4E-3), which would suggest that high HONO was not the cause of higher OH in the tropical plumes. Additionally, HONO that is initially emitted has only a transitory effect on OH, thus we do not discuss HONO as a major contributing factor to observed levels of OH in the Williams Fire.

7. Not much is mentioned regarding the relative timescales of the quick production of particulate ammonium and the slow/indistinct loss of ammonia as the plume ages, as shown in Figure 8. Could the large uncertainty in the gaseous NH3 at the source be a product of variable emissions compounded by quick conversion to particulate ammonium?

Authors Response. We want to emphasize that there is not a high uncertainty in the NH3 at the source, rather, the ΔNH3/ΔCO ratio for source samples increased from 0.0288 to 0.0577 in well-behaved monotonic fashion from the beginning to the end of the source sampling period, which lasted $\sim$2 h. It is only when this increase is all compressed to "time zero" on an aging plot that there is an apparent high variability. We attempted to relate each downwind ΔNH3/ΔCO ratio to the ΔNH3/ΔCO ratio at the calculated time of emission as an alternate analysis method, but it did not increase the certainty nor change the conclusion on amount of loss. We will rephrase this concept (P22505, L2) in a more detailed and clear explanation.

ACPD

11, C12503–C12518, 2011

8. It is clear that ΔOA/ΔCO2 decreased from 0.00355 to 0.00266 over the 4 hours since emission. However, I don't think that much can really be said about the initial decrease in the ΔOA/ΔCO2 ratio followed by a slow increase as shown in Figure 11. How was this fit/trend determined? It looks as if a simple exponential curve would also fit the same data points just as well.

Authors Response. While it is true that one function could also fit the data, two functions help us highlight the possibility of a turn-around in the ΔOA/ΔCO2 trend, which is both interesting and has theoretical underpinnings in the sense of POA volatility. It is also true that the OA aging sequence in many smog chamber studies (including many of those cited earlier in the paper) starts 45 min (lights out) after emission to allow characterization of the initial state – so it is useful to isolate the early period missed in those studies as a separate entity. We propose to keep the two trendlines, but better characterize them as an aid in pointing out a possible trend rather than as proof of a definite trend.

Specific and Technical Comments:

P22486 L13: Does the summary of BB in the "continental US" include Alaskan fires? If not, I would use the term "contiguous US." Authors Response. No, Alaskan fires were not meant to be included in this term. We will change "continental US" to "contiguous US" to clarify this.

P22495 L1: The term "nascent" should be clearly defined. I believe that the authors use this term to identify the freshest samples (0-10 min from emission) where it has been determined that little to no aging has occurred. Authors Response. We will add the definition of the term "nascent" at the beginning of Section 3.2 (the word means "coming into existence, or emerging"). Referee #1 is correct in that we use this term to define the freshest smoke samples that have undergone no measureable smoke aging. We think that the use of this term is appropriate, as it implies something just created, in the earliest stage of development.

ACPD

11, C12503–C12518, 2011

P22495 L9: The description of how the smoke age and estimated emission time were determined should be moved to the first paragraph of Sect 2.3 when describing the contents of Table 1. Authors Response. We incorporated the paragraph on estimated emission time into Sect 2.3. We agree that this information would be most helpful to the reader if it were found where we explain downwind sampling.

P22496 L27: Change “different time response of instruments” to “different time responses of the instruments.” Authors Response. This change has been implemented.

P22498 L19: OC is not defined in the text. Authors Response. We change “OC value” to “organic carbon (OC) value”.

P22500 L1-2: “: : :plume aging results were not obtained: : :” This wording is confusing. Authors Response. We rephrase this sentence to read: “Here we simply note that our plume aging results were obtained for a fire that had similar ERs, EFs, and MCE to those reported in the literature for other chaparral fires.”

P22500 L5-11: The brief comparison of the PM2.5 EF given in Sect 3.1 could be moved to Sect. 3.3 especially if Table 2 is removed. Authors Response. We agree that the PM2.5 EF data could work in either Section 3.1 or Section 3.3. However, we choose to keep this discussion in Section 3.1 as it addresses how representative the Williams Fire was of a typical chaparral fire.

P22500 L11: “Rather than emphasize late samples of smoldering combustion: : :” I am not sure what the authors are trying to say here. Does “emphasize” mean “measure” or is it referring to the relative weighting of certain data points? What does “late” refer to? If this sentence describes the sampling strategy, then it should be moved to Sect. 2.3. Authors Response. This sentence should stay in Section 3.1 as it discusses how representative our fire was of a chaparral fuel types. We suggest changing “Rather than emphasize late samples of smoldering combustion: : :” to the following: “Rather than measure samples of smoldering combustion acquired late in the fires development: : :”.

ACPD

11, C12503–C12518, 2011

P22500: Section 3.2 should be renamed “Plume evolution” in keeping with the title of the manuscript. Authors Response. We implemented this change.

P22500 L25-P22501 L4: The description of how the “smoke age” and “estimated emission time” were determined should be moved to the first paragraph of Sect 2.3 when describing the methods used and the contents of Table 1. Authors Response. See above response to P22495 L9.

P22501 L4-7: It would make more sense if the discussion of NH3 was moved to Sect 3.2.4. Additionally, I don’t follow the logic stated here. If you have greater uncertainty in the source emission ratio (presumably due to changes in emissions at the source), then one would expect that this would carry through to the downwind measurements (uncertainty = changes in source emissions + chemical and physical aging). For those species whose source emission ratios did not vary as much, then the differences downwind would reflect the changes due to chemical and physical aging (only). Authors Response. This is a minor point we hoped would explain some of the variability in ER and NEMR, along with the likelihood that these observed changes were in fact due to chemical and physical aging processes. To reduce confusion we will reword the original sentence: “Thus, with greater uncertainty in the initial value for NH3, differences between the downwind NEMR and the ER measured at the source are likely due to chemical and physical aging rather than changes in the emissions at the source.” We will replace this with: “Much of the decrease between the downwind NEMR and the ER occurs in the first hour following emission, suggesting rapid, initial NH3 removal by chemical and physical aging processes rather than changes in the emissions at the source”, as we were not trying to suggest that greater source variability would increase our confidence in downwind NEMR measurements (for NH3). We will move this text to Sect 3.2.4 where we go into depth on nitrogen-containing compounds.

P22501 L7-20: The authors discuss the nuances of Figures 6-12, which aren’t discussed until P22503, and before Figure 5 is referred to. In order to avoid unnecessary confusion and have the discussion follow a more logical progression, I suggest the fol-

ACPD

11, C12503–C12518, 2011

lowing: (a)Simply switch the placement of Sect 3.2.3 and Figure 7 regarding ozone with Sect 3.2.1 and Figure 5 regarding the alkenes and estimation of OH (b)The discussion and introduction of the aging plots and fits (currently L7-20) could now be included in Sect. 3.2.1 using the ozone plot as a direct reference in sequential order. Authors Response. We will try implementing this change and adopt it if improves the flow and organization of Section 3.2.

P22501 L21-L26: This paragraph includes an important discussion on the source ERs as a function of altitude and should be moved to Sect 3.1 (or later) when source ERs are first discussed. Authors Response. We agree that this information should be found where we discuss the ideal starting point for the aging plots, which is now in Section 3.2. We have reorganized the text so that the sole purpose of Section 3.1 is to compare the initial emissions of the Williams Fire with other chaparral fires (see response to Referee #1 Comment 1.2.3)

P22505 L2: There is enough scatter in both the source ER as well as the downwind NEMRs that the trend for NH3 is anything but "clear." Perhaps the ΔNH3/ΔCO aging plot mentioned shows a more distinct decay? Authors Response. The ΔNH3/ΔCO aging plot shows a similar decay to that of Fig. 8a. We will change the word "clear" to "implied" here.

P22508 L12: Figure 9b does not show ΔPAN/ΔNOx. Authors Response. PAN and NOx are both normalized to same sum and can thus be compared from the graph. We replace the following sentence: Figure 9b shows that ΔPAN/ΔNOx increased from $\sim$2.2% to $\sim$122% as the smoke aged $\sim$4 h." We add the following sentence instead: "Figure 9b shows the fraction of measured NOy species contained as PAN increases from $\sim$0.06 to $\sim$0.50, while the fraction of measured NOy as NOx decreased from $\sim$0.89 to $\sim$0.40 as the smoke aged approximately 4 h."

P22509 L21: It should be clearly noted that Cl atom has an appreciable rate coefficient with C2H2 so that the presence of Cl atoms in the plume would result in a decrease

ACPD

11, C12503–C12518, 2011

Interactive Comment

in ΔC2H2/ΔCO. The fact that the ratio didn't change may also give some indication that there was minimal mixing of the plume with background air. Authors Response. We will revise the following sentence "The fate of the chloride released from the particles is unknown, but it is unlikely that most of the released chloride constitutes a rapid source of Cl atoms as ΔC2H2/ΔCO did not vary over 4.5 h of aging (Tabazadeh et al., 2004)" We will replace the latter with the following text: "The fate of the chloride released from the particles is unknown, but it is unlikely that most of the released chloride constitutes a rapid source of Cl atoms. The Cl atom has an appreciable rate coefficient with C2H2 (Tabazadeh et al., 2004), and we did not observe a change in ΔC2H2/ΔCO over 4.5 h time since emission." The idea that no change in the ΔC2H2/ΔCO ratio suggests minimal plume mixing with background air may be true, but we did not measure ΔC2H2/ΔCO in background air and would prefer not to make this indication.

P22514 L2: It is more accurate to simply state that ∼50% of the initially emitted rBC particles are "thickly coated" as shown in Figure 12. The remaining 50% is classified as either "thinly coated" or "not coated" at the time of emission. Authors Response. The following sentence: "Figure 12 shows that most of the initially emitted rBC particles were, at most, thinly coated by other substances" will be replaced with: "At most ∼50% of the initially emitted rBC particles are "thickly coated" as shown in Figure 12. The remainder are classified as either "thinly coated" or "not coated" at the time of emission."

P22516 L16: It is sufficient to say that the plume was photochemically active. Authors Response. We changed "very photochemically active" to "photochemically active."

P22533: Remove comma between "a.m.s.l, (purple)" Authors Response. This change has been implemented.

P22537 Figure 5 caption: k1 and k2 are not defined in the text. These should be changed to kethylene and kCO to match the discussion in text. Authors Response. k1 and k2 have been changed to kethylene and kCO, respectively.

P22538 Figure 6 caption: The text "with plume aging (h)" should be changed to "esti-

ACPD
11, C12503–C12518, 2011

mated smoke age (hr)" to be consistent with the figure and subsequent figure captions. It should be clearly stated that the blue markers and error bars are the mean and standard deviation of the 10 source samples. The error bars on the black and red points at t=4.5 should be defined here. Authors Response. Thank you for these suggestions. We will change "with plume aging (h)" to "time since emission (h)." We will also define ERs and standard deviation markers clearly in the figure caption as needed.

P22539 Figure 7 caption: Is the y-intercept from Table 2 (slope-derived ER) or is it the mean? Authors Response. The y-intercept for O3 only (not an initial emission) is the mean initial NEMR, which is not shown in Table 2. We will change the figure caption to read as follows: "ΔO3/ΔCO vs. time since emission (h). The y-intercept was forced to the negative initial mean NEMR of the ten source measurements (blue circle). The standard deviation of these ten measurements is also shown (blue error bars). The error bar shown on the downwind sample is an estimate of the uncertainty in individual downwind samples."

P22540 Figure 8 caption: The error bars for data at t=4h is not defined. Authors Response. The downwind error bars are defined in the text, but we will define again here in the captions for clarity. The following sentence will be appended to the figure caption: "Error bars shown on downwind samples are an estimate of the uncertainty in individual downwind samples."

P22541: The blue points representing the source averages should be on top of the individual measurements so that you can see the mean s.d. Again, the error bars for the black and green data points are not defined in the caption. The abbreviated unit for hours should be consistent throughout the text, figure captions and axis labels. The figures have (hr) and some of the text refers to (h). Authors Response. We will standardize the abbreviation for "hours" as "h". We will also add a sentence explaining the downwind error bars (see above response)

P22542 Figure 10c: The blue data point should be on top. Authors Response. We will

ACPD

11, C12503–C12518, 2011

Interactive Comment

either make the blue data point larger, or make sure this data point will overlay other source points.

P22543 Figure 11: There are no blue data points showing the source mean ER for either parameter. Authors Response. Thank you, we will add the initial ER to this figure.

Anonymous Referee #2 R2.1. This manuscript presents analysis of the aircraft measurements of the chaparral fire that took place in California in November 2009. The measurements of chaparral fires are sparse and more research is needed to quantify the resulting emissions and physical and chemical processes in the plumes in order to understand the impact of these fires on atmospheric chemistry and composition. The authors describe pseudo-Lagrangian observations of multiple gaseous and particulate species sampled in the biomass burning plume from the time of emission, near the fire source, and up to 4.5 hours later, in the aged smoke downwind. The fire smoke was sampled in a relatively unperturbed environment, and therefore this experiment represents a valuable opportunity to study the photochemical processes in the plume as it is aging. The presented material is relevant to ACP and can be of great interest to its readers. I recommend this manuscript for publication after a few minor suggestions are addressed.

Specific Comments.

1. Page 22497, Lines 17-18. How did you decide which samples to include into the derivation of the initial fire ER? What "signs of aging" in the plumes at the distance of more than 1.8 km away you are referring to? Authors Response. We had to establish a cutoff for what we would consider a "source samples." During sampling, certain flight patterns were geared towards collecting samples as close to the source as possible. All of these samples were collected 1.8 km or less from the actual fire, thus we used this distance as our cutoff distance. One of the "signs of aging" that we are referring to is ozone formation. Only (and all) "source" samples (those collected $\leq$ 1.8 km from the

ACPD

11, C12503–C12518, 2011

fire) exhibit a negative ΔO3/ΔCO emission ratio, while all "downwind" samples exhibit a positive value. A negative ΔO3/ΔCO is explained from the reaction of entrained, background ozone with NO in the fresh plume that quickly titrates ozone to levels below background. In what we call "downwind" samples, the ozone formation has dominated ozone destruction long enough to yield positive ΔO3/ΔCO NEMRs. We will revise the following sentence to clarify this point: "Smoke samples collected more than 1.8 km from the source showed signs of aging (O3 formation) and were not used to derive the initial ER."

2. 22500, Line 25. In order to estimate the age of a smoke sample, did you use the windspeed that was averaged over the time period characteristic of travelling time from the source? This is currently not clear from the text. Authors Response. The time period characteristic of the traveling time from the source is what we are calling the "smoke age." Thus, we estimated the smoke age using the altitude to derive an average windspeed, since most of the transport occurred at the sampled altitude. Additionally, the windspeed was not that variable after ∼12:00 LT during the second flight when most of the aged samples were collected (Fig. 1c).

3. Page 22499, Line 4. Currently it is not obvious what the standard error of ER refers to, before it is mentioned later in the text (lines 16-17). Please describe this earlier in the text. Authors Response. We will add "of the slope-based ER" after "standard error" here, if this sentence is retained. As mentioned in the response to Referee #1, we plan on eliminating this table but will keep this point in mind when explaining and introducing other table parameters.

4. Table 2: the ER for NH3 to CO2, and ER for ozone are missing in Table 2. Authors Response. The NH3 ER is shown relative to CO, and ozone (like PAN) does not really have an ER as it is solely a secondary product.

5. Figures 6-12. Blue dots for ER are poorly seen (e.g. see Fig. 9a). Also, not all captions explain error bars for initial ERs. Explanation for the error bars for measurements

ACPD
11, C12503–C12518, 2011

downwind are missing in all captions. Authors Response. See Author Response to Referee #1 (P22541).

Anonymous Referee #3 R3.1. The manuscript presents a detailed analysis of the changes in the chemical composition of a biomass burning plume. The authors use the in-situ measurements of particles and gases made during a prescribed fire in California in November 2009. The plume was unperturbed by other pollution and particularly isolated and stable. As a consequence, using a pseudo-lagrangian approach, this case study allows a detailed discussion of the fate of the different particles and gases within a fire plume aging over 4.5h in the continental U.S.. Measurements made near the source are used to determine emission ratios and emissions factors. The evolution of the primary emitted and secondary produced compounds is discussed and compared to previous studies. The results of this experiment are valuable for further investigation and evaluation of model capability to reproduce such chemical composition changes in biomass burning plume. As a consequence, the results presented in this paper are of significance for a number of readers of ACP and the topic is relevant for publication in ACP. I recommend publication after revision.

General comments:

1. In Section 3.1, the authors discuss the error and uncertainties for the ERs and the NEMRs while the figures and example of NEMRs are not yet shown. In this Section 3.1, Table 2 is introduced but little discussion is given as the authors refer the reader to previous paper (i.e. Burling, 2011). As a result, it is difficult for the reader to find the objective of this section. If the authors' purpose is to discuss the difference between the estimated ERs (plot based) and the average of initial NEMRs, then the values for the latter should be added in Tab. 2. Also it seems that the initial NEMR values are expected in Table 2 as stated in the legend of Figure 7. Authors Response. This section had one main objective, which was to show that this was a typical chaparral fire in terms of ER, EF, and MCE, etc. We have renamed this section "Initial Emissions Compared to Other Chaparral Fires" to emphasize this. We have also moved all discussion on the

ACPD
11, C12503–C12518, 2011

starting point for the aging sequence and all NEMR/aging/uncertainty discussions to Sect. 3.2 in response to Referee #1 and your comment below.

2. The discussion about the ER standard error and the changes in NEMRs during the plume aging should be done in the Section 3.2. Having this discussion here in the text is misleading to the reader, as he expects here a discussion about the initial conditions and emissions ratios, factors. This discussion should be lead in each subsection, to determine if there was a significant change in the chemical composition of the plume downwind. Authors Response. This is an excellent suggestion, and we have renamed and reorganized Sections 3.1 and 3.2 to focus on initial emissions compared to other chaparral fires (Sect. 3.1) and plume evolution (Sect. 3.2), which gets into ER standard error and changes in NEMRs during plume aging.

3. It is not clear if the blue points and error bars in Fig 6 to 12 are for ERs or the average of the ten initial NEMRs and how systematic is the choice made between the two values (and why?) (see Fig. 8 for example). Adding a sentence of comment about this point in the beginning of each section will add some clarity. Authors Response. We emphasize why the slope-based ER is the best representation of an emission ratio (rather than the mean initial ER for the ten source samples) in Section 2.4.2. Additionally, in Sect. 3.2 we will clarify that the standard deviation of the mean of the ten initial NEMRs is the best estimate of the uncertainty in the starting value for individual downwind samples.

Specific comments: Figure S1. The time of the profile is not specified in the caption of the Figure nor in the text. Could the authors specify it in both of them? How does the BL height changed between the two flights and during the experiment? Authors Response. Yes, we will add the time the profile was taken to both the text and the Figure caption. Unfortunately we did not measure the BL height in the second flight.

p22494 l4-5 The profiles of the measured wind direction are not shown on Fig1.c. Giving those profiles beside the profiles of the wind speed will support the statement. Authors Response. The plot was too cluttered when the wind direction data was over-

ACPD

11, C12503–C12518, 2011

laid on Fig. 1c. We could possibly add a second supplementary figure, but the wind direction data we have is from close to the source where the air and winds were turbulent. At this point, we don't believe we have enough confirming data to make a clean and easily-interpretable figure, but the data are consistent. p22494 l8 The authors state that the mean wind speed in the lower layer is roughly 2.5m/s. Is that an average between the two flights? Please clarify. Authors Response. Yes, this average of 2.5 m/s is the average windspeed between the two flights measured at altitudes less than 1200 m a.m.s.l. We will clarify this in the text.

p22495 l1 The authors use the term "nascent" which is not clearly defined and not of common use. Do the authors mean they sample the freshest plumes? Are these locations above the fire area? Also the authors state that the center and top portions were sampled. Was it done systematically? Or some of the samples are from the center and others from the top portions? Authors Response. Regarding the term "nascent," please see the response to Referee #1 (P22495 L1). Yes, we tried to systematically sample different altitudes within the nascent plume (updraft core).

p22506 l18 Please introduce Fig 9 at the beginning of the paragraph. As a general comment, it is much more easier for the reader to follow the discussion if the figures are clearly introduced at the beginning of the paragraph it may concern, which is often not the text in the submitted version of the manuscript. Authors Response. This first paragraph discusses the general significance and Figure 9 is introduced as soon as we get into discussing the actual data.

p22508 l7 Please specify that the plot showing the change in PAN versus CO2 is not shown for clarity purpose. Authors Response. Thank you, we will append "(figure not shown)" to the following sentence: "Overall, ΔPAN/ΔCO2 grows by a factor of 10.5 $\pm$ 5.2 in 4 h aging and accounts for 22$\pm$6% of the initial NOx on a molar basis."

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 22483, 2011.