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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Boreal forest fire emissions in fresh Canadian smoke plumes: C_1–C_{10} volatile organic compounds (VOCs), CO_2, CO, NO_2, NO, HCN and CH_3CN" *by* I. J. Simpson et al.

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1. This paper presents result from the 2008 ARCTAS B mission component that measured boreal fire emissions, presenting emission data for a number of compounds for the first time. The paper is well written and the measurements make a very useful addition to the literature. I recommend publication of this paper following the consideration of some minor comments, given below.

- We thank Reviewer 2 for these comments.

2. Mixing of background air with the BB plumes could affect the calculated ERs, since the ratios of compounds to CO are often different in the background compared to the plume. Could the authors consider this effect and comment of its impact on their analysis?

- This is a good point and we have discussed the possible corruption of the calculated ERs as a result of background air mixing with the biomass burning plumes. If a plume is diluted with background air, $\Delta X/\Delta Y$ is preserved as long as the background air is constant in X and Y. However caution is needed because once X/Y in the background air changes, subsequent $\Delta X/\Delta Y$ no longer reflect the "exact" original characteristics of the plume. For diluted aged plumes, this effect can potentially be significant. One way to test for this effect is using the modified combustion efficiency, where MCE = ΔCO2 / (ΔCO2 + ΔCO). Since by definition MCE cannot be > 1 at the source of a fire, an MCE > 1 means that ΔCO2 is a small negative number. To cause MCE > 1, CO2/CO would have to change in the background air during transport, with X/CO likely changing as well, where X is a VOC. Here our average MCE for Plumes 1-5 ranged from 0.71-0.95 (i.e. < 1), suggesting that any impact from changing background values was likely modest. In addition, we selected fresh plumes and our method of calculating ERs–which subtracts off the background mixing ratios and forces the fit through zero–effectively weights the ERs to high values that are minimally affected by background issues. Finally, our values for those species that have been measured previously are in good agreement with measurements in very fresh boundary layer biomass burning plumes and laboratory biomass burning fires (e.g., Yokelson et al., 2008; Akagi et al., 2011), both cases in which changing background concentrations is not a complicating factor. Therefore our best assessment is that non-constant background air mixing with the biomass burning plumes did not have a substantial impact on our results.

To address this comment we have added the following text on P9531 L28: "Calculated ERs can potentially be affected by biomass burning plumes mixing with non-constant background air, which can have different ratios of compounds to CO. Our method of calculating ERs–which subtracts off the background mixing ratios and forces the fit through zero–effectively weights the ERs to high values that are minimally affected by background issues. In addition, our ER and MCE values are in good agreement with measurements in very fresh boundary layer biomass burning plumes and laboratory biomass burning fires (e.g., Yokelson et al., 2008; Akagi et al., 2011), both cases in which changing background concentrations is not a complicating factor. This gives some indication that we were successful in selecting relatively fresh plumes where the original signature was still clear."

3. Another possible effect in these studies is fire-induced convection, which would increase vertical transport of boundary layer air in the plumes relative to the mean transport that the un-perturbed BL experiences. Could this be the reason behind the slightly higher values for HCF-141b in plumes, and the slight apparent negative CH3CCl3 emissions in this study and the previous Australian study?

- We agree that boundary layer air can get entrained in plumes because of fire-induced convection, which can potentially impact the results when a strong vertical gradient is present. In this study, the slightly higher HCFC-141b values in the plumes would require entrainment of enhanced BL air, while the lower CH3CCl3 values in the plumes would require entrainment of depleted BL air. It seems unlikely to get the opposite effect for two anthropogenic products emitted into the BL, but we investigated this further by looking at the vertical gradients of HCFC-141b and CH3CCl3 in the smoke samples and boundary layer air samples collected during the five plume encounters of Flights 17-19. The five smoke plumes and their corresponding background air were sampled at altitudes between 0.7-5.4 km (Table 1), with average sampling altitudes of 2.1 $\pm$ 0.2 and 2.0 $\pm$ 0.4 km, respectively. At these relatively low altitudes the vertical gradients of HCFC-141b and CH3CCl3 were not statistically significant. The average HCFC-141b mixing ratios between 0-1 km, 1-2 km, and 2-3 km were 21.05 $\pm$ 0.85 pptv, 21.14 $\pm$ 1.07 pptv and 21.31 $\pm$ 0.73 pptv, respectively, and the average CH3CCl3 values were 12.46 $\pm$ 0.08 pptv, 12.44 $\pm$ 0.12 pptv and 12.29 $\pm$ 0.16 pptv, respectively. When

treated separately, the in-plume and background samples also did not show significant vertical gradients (not shown). Therefore we did not find evidence for HCFC-141b enhancement and CH3CCl3 depletion in the BL, and we conclude that fire-induced convection did not affect the measured CH3CCl3 and HCFC-141b mixing ratios in the plumes.

To address this comment we have added the following to the text at P9533 L6: "HCFC-141b was measured using GC/MS and the reason for its slight enhancement in the plumes is unclear, but we found no evidence that fire-induced convection was a contributing factor."

4. Pg 9529, Line 17. I realize this is a standard quantity, but the Modified Combustion Efficiency" should be defined here.

- We have expanded the definition of MCE on P9529 L15-17 to the following:

"Modified combustion efficiency (MCE), defined as ΔCO2 / (ΔCO2 + ΔCO), is an indicator of a fire's combustion stage, ranging from near 0.80 for smoldering combustion to 0.99 for pure flaming combustion (Akagi et al., 2011)."

5. References: "an der Werf" should be "van der Werf"

- Thank you. We have made this correction.

6. Figure 2: The text in the panels is a bit hard to read.

- We have enlarged the text in the panels.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 9515, 2011.