

***Interactive comment on “A review of biomass burning emissions, part I: gaseous emissions of carbon monoxide, methane, volatile organic compounds, and nitrogen containing compounds” by R. Koppmann et al.***

**Anonymous Referee #2**

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General Comments

This paper provides an extensive collection of numerical data describing emissions of trace gases from biomass burning. As a review paper, its focus should not only be to present all of the data, but to also provide conclusions based on the collection of data, not just discussions about selected sets of results (see next comment). In its current organizational structure, the sections (and tables) for different molecules attempt to describe wide ranges of observations, often without differentiating between

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the different types of fires (which predominantly dictate the emission ratios). This paper would be far more valuable if sections (and tables) were organized by fire type and if conclusions were drawn about emission ratios for each of the different types of fires. Currently, the reader is immersed in a sea of numbers in tables, some of which are reiterated in the text, but is left to sift through all the numbers to draw his/her own conclusions about trace gas emissions from biomass fires.

The detailed descriptions of some experimental methods and results in the text appear to favor a few selected studies. What do these detailed descriptions contribute to the paper? To be fair, every study with results listed in the tables should be described in the same amount of detail, or none of the studies should be described in detail. A review paper should not give preferential treatment to a handful of selected studies unless they provide something unique or beyond the scope of the other studies.

Sections describing stable isotope studies and ozone production from VOCs are not really within the framework of this review paper. They tend to detract from the primary focus.

The paper contains many typographical errors, citation/reference errors, and in numerous instances, poor sentence structure. It appears to have been hastily prepared for publication, likely without proofreading and certainly without the use of a spelling checker. These errors greatly increased the amount of work necessary to review the paper and are unacceptable in a manuscript submitted for peer review.

### Specific Comments

Abstract L6-7 "8700 Tg of dry matter/year are burnt each year in total". Does "in total" mean "globally" ?

Abstract L25 what does "our results" refer to? Global emission estimates?

10459 L8 cellulose and hemicellulose are both carbohydrates and polysaccharides

10461 L14-16 This sentence requires further explanation that oxidized gas emissions

correlate best with CO<sub>2</sub> emissions and reduced gas emissions correlate best with CO emissions, as demonstrated by Lobert et al., 1991. Otherwise, it is not clear why CO is a surrogate for hydrocarbons and particulate carbon.

10461 L25 This equation pertains only to emissions of carbonaceous gases, and this must be mentioned.

10462 L6-7  $C_f$  can only be measured post-fire if  $M_f$  was measured pre-fire.

10462 L16 Here your symbols  $[n]$  and  $[C]x$  depict the mass concentrations of  $n$  and Carbon (as species  $x$ ) emitted in fire plumes. Later on (10464 L22) your definition of emission ratio includes  $\Delta[x]$  to indicate mixing ratio enhancements in plumes, so  $[x]$  is now the molar mixing ratio of  $x$ . This is very confusing to the reader - do each of equations 2-7 depict excess mass concentrations in the plume? If equation 4 is transformed into equation 7, why does  $[C]x$  in equation 4 change to  $[x]$  in equation 7, and shouldn't  $[x]$  in equation 7 really be  $\Delta[x]$  as defined in equation 8? I feel that the concept of emission ratios should be introduced before emission factors and combustion efficiency, and that different symbols should be employed for molar mixing ratios and mass concentration enhancements in plumes. This will facilitate a discussion of how emission ratios can be converted to emission factors if the primary carbonaceous gas and particle emissions were measured and the fuel C or N content is known.

10465 L11-15 After mentioning that estimates of biomass burned differ by as much as a factor of 4, you describe the estimates of Andreae (1991) like they are the ones we should believe (and you don't even mention what the other estimates are). The reader is not told whether the Andreae (1991) estimate is at the high or low end of the factor of 4 range of estimates. This is a highly biased presentation of the information available in the literature and is not suitable for a review paper.

10468 L21-23 Some clarification is needed: Aren't NMHC and oxygenated NMHC together classified as VOCs? Your terminology needs to be consistent throughout the paper. VOC is used exclusively until Section 5.3 entitled "NMHC" discusses VOC and

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NMHC like they are exactly the same.

10468 L23-26 Don't several of the Cofer et al. papers present NMHC emission ratios for temperate and boreal forests?

10469 L1-3 It should be mentioned again that reduced gases are emitted along with CO in the smoldering phase of fires, and hence their emissions are better correlated with emitted CO than with emitted CO<sub>2</sub>. Then it should be mentioned that your choice of presenting only emission ratios to CO<sub>2</sub> makes the intercomparison of emission ratios for reduced gases less conclusive.

10469 L20 Symbols for excess mixing ratios of x have now changed to  $\Delta(x)$  instead of  $\Delta[x]$  ?

10470 L1-16 This represents a prime example of how the reader is immersed in a sea of numbers for CO emission ratios, with seemingly no conclusions presented about CO emission ratio averages or ranges for each of the different fire types. Summarizing the published CO emission ratios for different fire types would provide much more valuable information.

10471 L4 "rather long lifetime of CO in the troposphere" is a very qualitative statement that should be made quantitative. The same goes for "Methane is not a very reactive compound" in L21.

10472 L11-16 why do you go into great detail about observations of methane mixing ratios over Brazil being above background levels when these observations are not connected to an emission ratio?

10472 L20-21 The ICFME acronym and location were already described (10470 L4-6)

10472 L23-25 Why is it "even more important to know the emission factors" for methane "for an up-scaling to global emission rate" ?

10473 L16-26 Why so much detail about the Greenberg et al., 1984 study and their

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calibration technique? Why re-iterate the emission ratio data that are already presented in Table 4?

10477 L6-13 You are very critical of the emission factors that were calculated from published emission ratios by Andreae and Merlet (2001), implying that large errors are introduced by their procedure. But, you fail to address that your compilation of only emission ratios to CO<sub>2</sub>, if these data are used to estimate emissions, will also propagate large errors, especially for trace gas emissions that are poorly correlated with CO<sub>2</sub> emissions.

10478 L15-20 FTIR measurements by Hurst et al., 1994 were not made in situ, and emission ratios for HCN and CH<sub>3</sub>CN were presented along with HCHO and CH<sub>3</sub>CHO.

10479 L9-12 Halogenated hydrocarbons emitted by biomass fires include CH<sub>3</sub>Br and CH<sub>3</sub>I, so why do these emissions depend “to a large extent on the chlorine content of the fuel”? By how much did the Lobert et al., 1999 data for Cl content of fuel vary?

10480 L12-18 Why is there no mention of CH<sub>3</sub>Br emissions presented by Mano and Andreae, 1994?

10480 L28-29 What “number of halocarbons is rapidly declining”? CH<sub>3</sub>CCl<sub>3</sub> and CH<sub>3</sub>Br? Others? These hardly constitute “a number of halocarbons”.

Section 5.6 There needs to be some information presented here about emissions of N-containing gases being dependent on fuel N content. Comparisons of NO<sub>x</sub>/CO<sub>2</sub>, N<sub>2</sub>O/CO<sub>2</sub>, etc. are virtually meaningless without knowledge of the N/C ratio of the fuel. Lobert et al., 1991 found a wide range in the N content of different biomass types (0.15-2.4% of dry weight) and presented evidence that NO<sub>x</sub>/CO<sub>2</sub> and N<sub>2</sub>O/CO<sub>2</sub> were proportional to the molar N/C ratios of the fuel.

10482 L24 “All values” contradicts “two compounds differed from the general observations” (10483 L1-2)

10483 L9-11 Wouldn't CH<sub>3</sub>Cl also be produced by radical recombination in the flame

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since chlorine is tied up as chloride ion in the biomass?

Section 8: Your intent for global emissions to not be the focus of the paper is evident in statements like: “seem to be something like a lower and upper limit” (10486 L25) and “seems to be on the order of” (10487 L4-5)

10486 L13-16 You need to describe how the values in Table 7 were obtained. How was the CO emission ratio for each fire type determined from the numerous published data sets?

10487 L8 500 Tg / 2780 Tg = 18%, not “about 25%”

10487 L22-23 how are CO emissions of 375-400 Tg estimated in 2004 “somewhat higher than previously thought” when previous estimates were 500 Tg?

10488 L10-12 Are there really “large uncertainties in the emission data” or is there simply natural variability in emission ratios measured in different locations, under different fire conditions, at different times? This statement is so negative it implies that all we have is a collection of really bad measurements, which is definitely not the case.

10488 L20 “the” trend implies that there is a trend. Please change to “a” trend.

10489 L26-30 The CH<sub>3</sub>Cl budget in WMO, 2003 depicts 900 Gg from biomass burning and 3000 Gg total sources = 30% (not 20%). Biomass burning is also an important source of CH<sub>3</sub>Br (20 Gg out of 160 Gg, WMO, 2003), and estimates in Rudolph et al., 2000 for biomass burning emissions of CH<sub>3</sub>CCl<sub>3</sub> range as high as 12 Gg per year (industrial emissions were 40 Gg in 2003). Biomass burning emissions of these halogenated compounds are of global importance.

10491 L1-4 Your portrayal of problems with NO<sub>x</sub> and O<sub>3</sub> measurements needs support from at least one reference.

10491 L5-10 What is more important to understanding the impacts of biomass burning emissions on regional atmospheric chemistry, monitoring emissions at the fire or mon-

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itoring emissions slightly downwind where longer-lived secondary products have been formed?

### Technical Corrections

There are too many occurrences of poor word choice, poor grammar, and spelling errors to even begin to list them all. Here is a very small subset:

10458 L15 “The biomass combustion processes and its relation” 10470 L6 “North West Territories” should be Northwest Territories (Table 1 says “New Territories” 10470 L15 “founr by Brocard” 10471 L16 “satallite products” 10472 L7 “FOS/DEFACE” 10475 L10-12 “Especially the response of fires to wind, etc.” is not a compete sentence. 10476 L11 “inmvestigated”

I suggest the authors engage in a serious proofread of this manuscript.

There are table elements missing: For example: Table 1: No Time of Investigation is given for EUSTACH or for ICFME. The latter was June-July 1997.

Table 2. No Comments are given for Andreae et al., 1988 (Amazon Basin) or Hurst et al., 1994 (Australian savanna). This table and tables 3-6 should be arranged by fire type, not publication date.

Table 3. Comments are missing for several studies.

Table 6. Comments (fire types) are missing. Data are missing - e.g.,  $N_2O/CO_2$  from Lobert et al., 1991;  $NH_3/CO_2$  and  $N_2O/CO_2$  from Hurst et al., 1994. Values for  $NO_x/CO_2$  are not within the range (0.5-3.5%) given in the text (10481 L9).

Table 7. Carbon released (42 Tg) cannot be greater than dry matter burned (20 Tg)

This review paper is a gold mine of references for scientists wanting to know more about gaseous emissions of biomass fires. For this reason, it is important that citations and references be correct. There are many instances where citations in the text do not match references at the end, in both text and tables. Examples:

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Greenberg and Zimmerman 1984 in text; Greenberg et al. 1984 in refs  
Andreae et al. 1997 in text; Andreae 1997 in refs  
Lobert and Warnatz 1993 in text; Lobert and Warnatz 1996 in refs  
Andreae et al. 1991 in text; Andreae 1991 in refs  
Levine 1996a in text; Levine 1996 in back  
Andreae et al. 1996 - needs a, b, or c (there are three of these in the reference section)  
Andreae et al. 1993 in text; Andreae 1993 in refs  
van der Werf 2004 in text; Van der Werf et al. 2004 in refs  
Ward 1991 in text; Ward 1990 in refs  
Czapiewski 1999 in text; v. Czapiewski 1999 in refs  
Hurst et al. 1994a, b in text; Hurst et al. 1994 and 1996 in refs  
Lacaux et al. 1992 in text; Lacaux et al. 1993 and 1995 in refs (1995 appears 2x in refs)  
Lindsay et al. 1996 in text; Lindsay et al. 1996 in refs  
Hurst et al. 1993 in text; Hurst et al. 1994 and 1996 in refs  
Helas et al. 1995 - needs a, b, or c (there are three of these in the reference section)  
O'Malley et al. 1997 is missing title of paper  
Thompson et al. 1996 - needs a or b (there are two of these in the reference section)  
This is not an exhaustive list of citation/reference problems, only examples!

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 10455, 2005.

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