

## ***Interactive comment on “Primary and secondary sources of formaldehyde in urban atmospheres: Houston Texas region” by D. D. Parrish et al.***

**Anonymous Referee #3**

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The paper provides a very interesting and elaborate contribution to improve the estimate of the relative roles of primary emission of formaldehyde and its photochemical formation from anthropogenic VOCs (alkenes) in the highly industrialized Houston Texas region. The authors rely on measured data from several field studies and on the measurement-constrained EPA NEI 2005 inventory to determine the primary emission rate of formaldehyde and its secondary production rate from ethene and propene oxidation by OH. Secondary formation is identified to be the predominant source of formaldehyde with about 95% contribution. The relative role of primary emission (5%) stands in contrast to findings from previous investigations that estimated a fraction of up to 50% by direct emission. The paper discusses possible reasons for the different results and provides convincing arguments why primary emissions were likely overestimated in previous studies. The paper is well written and is suitable for publication in  
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ACP after the following revisions have been made.

(1) The concept that is used for the estimation of emission and production rates of HCHO is based on various assumptions that introduce uncertainties in the calculated results. The assumptions should be explicitly listed and the related uncertainties quantitatively estimated. For example, a constant yield of HCHO from the oxidation of ethene and propene has been assumed. How much variability is expected for the yield which may depend on solar radiation, NO<sub>x</sub> levels etc. How much uncertainty does the assumption of a fixed yield introduce into the estimated secondary production rate? On page 32610 (lines 2-4) ozone and nitrate radicals are mentioned as other possible oxidants that degrade alkenes. How large is their likely contribution to the secondary production of HCHO?

(2) The estimated uncertainties of the primary emissions (30%; Fig. 4) and the secondary formaldehyde flux (40%; page 32610, line 12) need explanation. How were these values derived? The percentage contribution of the secondary formaldehyde production to the total rate is given as 95% (Table 4). Why is the uncertainty of this fraction so small (+/-3%)? Is this a typo?

(3) Although it is not the focus of the paper, it would round off the discussion if the authors provide a brief statement how much formaldehyde may be contributed by biogenic VOCs in the Houston Texas region compared to anthropogenic sources.

Other comments:

- page 32616, line 27: give examples for other possible sources. Do ships play a role?
- Fig. 2: was the intercept of the regression lines constrained to the measured background concentrations of ozone and formaldehyde, or were two parameter fits (slope and intercept) applied to the data pairs?
- Different units (kg/h and kmole/h) are used in the paper. For consistency, I suggest to use only kmole/h throughout the paper. That would make it easier to compare numbers

and simplify the tables.

- Tables 1, 3 and 4 are awkward to read. Rates should be given consistently in the same unit (kmole/h). The meaning of the error bars should be specified in all tables (are these 95% confidence intervals?). The percentage values in parentheses (Table 4) need to be explained. I guess these are relative contributions to the total (primary + secondary) rate?

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 32601, 2011.

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