

## ***Interactive comment on “Temporal and spatial variations in rainwater methanol” by J. D. Felix et al.***

### **Anonymous Referee #1**

Received and published: 31 January 2014

Referee's Comments for J.D. Felix, S.B. Jones, G.B Avery, J.D. Willey, R. N. Mead, and R. J. Kieber, Temporal and Spatial Variations in Rainwater Methanol, Atmos. Chem. Phys. Discuss, 14, 1375-1398, 2014.

#### General Comments

This paper provides a summary of the first long-term study of methanol concentrations in rainwater. This summary of the methanol observations may be of aid to modellers in constraining the magnitude of the wet deposition sink of methanol in the temperate Northern Hemisphere. The methodology of this study is carefully presented and the results are complemented by well chosen and well selected Figures. However, in my opinion, the paper will require substantial revision before it can be considered for pub-

C38

lication. The authors need to give much more thought about the interpretation of their data and how that can be most effectively communicated to the readers. For the benefit of the authors, I have included a number of Specific Comments primarily related to the Introduction, Discussion and Conclusions. There is also a list of Technical Comments to assist the authors in improving the readability and clarity of the paper.

#### Specific Comments

1. Abstract: (a) The first sentence of the Abstract is probably correct in stating that this study is the “first detailed analysis”, but it is not the first to measure ‘methanol concentrations in rainwater’, as noted later by the authors on page 1382, lines 5-6. (b) The sentence in lines 18-20 seems contradictory. Even if there is an ‘increase in biological activity’ leading to an enhanced production of methanol, this does not infer a ‘direct relationship to photochemical methanol production’. Also, an increase in production does not necessarily imply a direct relationship. (c) The meaning of ‘fluctuating methanol concentrations’ in lines 16-17 is not clear; please clarify. (d) Some other changes might be necessary in the Abstract in response to General Comments later in this review;

2. Introduction: (a) The introduction, although informative, seems unnecessarily long and is focussed on aspects that seem only marginally related to the main theme of your paper. It is not clear from the objectives of your paper, why it is important to discuss methanol sources. (b) Given the main theme of the paper, I had expected more background information about the processes that control the concentration of methanol in rainwater, i.e. the physical and chemical processes involved in the entrainment of methanol in clouds and in rainwater. With this in mind, the authors might consider deleting at least the last sentence in paragraph 1, paragraph 2, and the first two sentences of paragraph 3. This would provide the opportunity to include material more relevant to the aqueous-phase.

3. Introduction, paragraph 3: (a) If the first part of this paragraph is retained, it will

C39

require reorganization and clarification. The word 'constrain' is not appropriate in the context of sentence 1 (line 26). Further, it is not clear how 'these efforts' could have led 'to wide discrepancies in global budgets' (lines 26-27). (b) Sentence 2 needs major revision. (c) The first part of sentence 3 is fine, but one can't have 'a paucity . . . of its role' .. revision is necessary. (d) In the final sentence, it is not clear to what 'this uncertainty' refers. If it refers to the lack of methanol concentrations in precipitation, it is not clear how this would lead to a narrower range of wet depositional sinks.

4. Section 2.4: (a) It might be better to include at least some of the last paragraph in this section at the beginning of the section so that the reader understands what is meant by 'supporting analyses'. (b) There is no mention of the method used for measuring acetaldehyde (referred to in Section 3.4). (c) What meteorological measurements (other than rainfall) were made and how were they measured? Given its importance to this study, it would be useful to include a Figure (line or bar chart) showing the times and magnitudes of the rainfall events during the study period. If temperature readings were not made as part of the study, you might consider using data from a near-by meteorological station; these could be shown in the same figure as the rainfall data.

5. Section 2.5: (a) I may be showing my ignorance about trajectory analysis here, but I wonder why you are tracking air masses rather than clouds, given that clouds are the source of aqueous-phase methanol? (b) In line 26, you refer to a 'county basemap'. International readers may not be familiar with land division into counties, or what is meant by a 'county basemap'. Presumably the basemap gives methanol emission fluxes, as shown in Figure 2. (c) Also, it would be useful if the units in Figure 2 were given in SI units, or a conversion factor included in the caption. (d) In line 27, is the inclusion of 'biogenic' necessary? It raises the question of how anthropogenic methanol trajectories are portrayed, or how you have distinguished biogenic methanol from anthropogenic methanol. How does the basemap distinguish biogenic methanol from anthropogenic methanol?

C40

6. Section 3 (lines 1-10): (a) It would be useful to include median concentrations (as well as means). Are the medians similar to the means? (b) There is a very wide range of methanol concentrations for the rain events (noted here and in Fig. 6), yet this variability is not discussed or explained in the paper. This would seem to be quite important? (c) lines: 6-10: In fairness to Snider and Dawson, they acknowledged the inexactness of their precipitation measurements in the Discussion of their paper. I agree that 'direct comparison to this earlier study should be made with caution', but I suggest that the last part of the sentence be revised.

7. Section 3.1, paragraph 1: (a) There are several problems here that need to be addressed. Firstly, is 'storm' synonymous with 'rain event', or different? The alternation of these words in this section is confusing for the reader. (b) Secondly, and more important, if standard deviations are considered (as they must be), there is no significant difference between the average terrestrial methanol concentration ( $1.5 \pm 0.5 \mu\text{M}$ ) and the average marine methanol concentration ( $1.1 \pm 0.2 \mu\text{M}$ ). Thus, the statement in lines 17-18 is questionable. To assist in this matter, a t-test should be applied to the data and the results included in the paper. (c) Also, if marine gas-phase concentrations are an order of magnitude less than terrestrial gas-phase concentrations, how can the marine concentration in this study be so large? I would suggest, without checking the sources, that these values are average value for the whole ocean, and may not be appropriate for comparison to concentrations near the coast. (d) Thirdly, in comparing gas-phase concentrations with aqueous-phase concentration, one has to be sure that they are in equilibrium and that they are at the same temperature. It is not clear that this is the case here. (e) Fourthly, in the last sentence of this paragraph, I don't think there is sufficient evidence from the concentrations given here to make this statement. (f) There is no problem in including the results of the air trajectory study; but how you interpret them needs to be reconsidered.

8. Section 3.1, paragraph 2: (a) It might be argued that coastal rain events have a smaller concentration of methanol than mixed samples; however, I suggest (partic-

C41

ularly considering the relatively small number of sample analysed) that there is little significant difference between the 4 types of rain events. The question to be posed in this paragraph is why? (1) It seems difficult to argue for an ocean source that is comparable to that on land, if marine gas-phase concentrations are on average an order or magnitude less than terrestrial gas-phase concentrations; (2) methanol can be transported from the continental to the marine atmosphere, but is that consistent with your trajectory analysis for marine and coastal rain events?; (3) local scavenging seems the most likely of the three explanations offered in this paragraph. But before offering an explanation, it is necessary to discuss the physical processes involved: (i) where and how does the scavenging occur; (ii) how stable is the cloud system from which the rain comes? (iii) how long is the air mass in contact with continental air?, and (iv) how long does it take for methanol to equilibrate with rain droplets?; or (v) is the methanol in the rain sample in equilibrium with the atmospheric methanol?

9. Section 3.2, paragraph 1: (a) The last part of the first sentence of this paragraph, following '(Fig. 3)', does not seem related to the first part of the sentence. Based on their titles, the studies of Kieber 2001a and 2002b, do not appear to include measurements 'at this location'. If the latter part of this sentence is retained, the cited studies should be compared with the results of the current study more fully later in the section. (b) It would seem better to compare the diurnal variation in methanol that you observed in the rain samples with the typical diurnal variation for gas-phase methanol (see paragraph 2). (c) The last sentence of paragraph 1 would be better included in the caption of Fig. 3.

10. Section 3.2, paragraph 2: (a) There is considerable confusion concerning the interpretation of the results depicted in Fig. 3. In sentence 2, it is not at all clear how you can conclude that the larger methanol concentration in period III was the result of photochemical production of methanol, after earlier in the paper stating that methanol production is 'primarily biogenic' – and also that there is a 'direct proportion'. (b) Also, are both of the production processes indicated in sentence 2 photochemical? ... and

C42

how much methanol are they likely to contribute? (c) The latter part of sentence 2 lacks clarity; the 'biogenic and anthropogenic activity' is not identified. Are you referring to biomass burning? It seems unlikely that increases in solvent evaporation, for example, could account for the increase in methanol concentration in period III. The authors should reconsider the inclusion of this sentence. (d) The third sentence of the paragraph is not well worded; I'm not sure what is meant by 'is consistent with', or what is meant by 'varying vegetation', or what you consider to be the cause of the 'light-stimulated release'. You might consider what explanation is provided by the referenced authors to account for the observed diurnal pattern in the gas-phase. (e) Whatever, the reason for the increase in the methanol concentration in period III, it is difficult to assess your arguments without a knowledge of the diurnal patterns of temperature and light intensity during the rain events considered here (it would be helpful if you could provide them). Also, it would be useful to indicate how many of the rain events in the various time periods originated from marine trajectories; did these exhibit similar diurnal patterns to methanol from a terrestrial trajectory? (f) It is important to remember that you are measuring methanol concentration in rainwater, not in the gas-phase of the atmosphere. Rain events in temperate regions are often accompanied by a decrease in temperature, which would increase the solubility of methanol in rain droplets leading to an increase in aqueous-phase methanol, independently of the gas-phase concentration.

11. Section 3.2, paragraph 3: (a) Again in this paragraph, you need to address factors that might lead to reduced methanol concentrations in rain samples collected at night (which are not necessarily the same factors that could affect gas-phase concentrations of methanol). For example, during a rain event, one might expect that wet deposition would exceed dry deposition. (b) It is not necessary, or wise, to list every possible mechanism for 'night-time decrease' that you can think of. Consider what is most likely to explain your results. It is unlikely that you would have 'dew formation' during a rain event. Advection of marine air may have an impact, but this should be easy to test from the air mass trajectories at the time of the rain events. It would also seem likely that if

C43

the leaf stomata of plants close at night, biogenic methanol emission would decrease or cease, leading to substantially lower night-time concentrations of gas-phase methanol. (c) Both paragraphs 2 and 3 of this section should be rewritten, with more thought given to factors influencing aqueous-phase methanol concentrations, and whether or not they should be similar to changes in gas-phase methanol.

12. Section 3.3: (a) Examining Fig. 4 suggests that there is no significant difference in methanol concentrations between the fall, spring and winter seasons, but that the methanol concentration in the summer season is significantly greater. Use a t-test as a guide to support or reject this hypothesis. (b) Your primary aim in this section should be to (i) summarize the observations; (ii) suggest reasons for seasonal similarities and differences. Do this in a simple and straight-forward manner. The chief observation is that there is an enhanced concentration of methanol in summer rain events. Discuss the reasons for this first. The second important observation is not that there is a smaller concentration of methanol in the rain samples during the other seasons, but rather that in these seasons the methanol concentration it is about a third of that during the summer – still a very significant concentration. Why? (c) It is recommended that this section be re-written to improve its organisation and clarity. As mentioned earlier, the inclusion of temperature data might be useful (particularly, since methanol emissions seem to be temperature dependent).

13. Section 3.3, paragraph 1: (a) Based on your observations and noted in the previous comment, there seems no reason to group the seasons into winter and fall in paragraph 1, and spring and summer in paragraph 2. (b) The discussion of the winter/fall results in paragraph 1 should come after the discussion of summer data, which is currently in paragraph 2. (c) Sentence 4, beginning on line 10, is poorly constructed and misleading. It may well be worth pointing out that in the four studies referenced in this sentence, winter concentrations of methanol in the atmosphere are about a 1/3 of those in the summer (you could even supply data from these studies for comparison); but, as currently worded, there seems to be a suggestion in this sentence that the re-

C44

sults of these 4 studies determine the results in your study. (d) lines 13-16: You need to have supporting evidence from your study to argue that the 'greater percent contribution' is anthropogenic. It is not reasonable to expect that the results of Hu's study will automatically apply to your study. (e) Sentence 6, beginning on line 16, is poorly constructed; if you had first noted that methanol emissions from plant decay is quite small, it would not 'be expected that fall concentrations would increase'. Also, note that the estimates for plant decay in the studies referenced here are global averages, and may not apply to your location. (f) The argument presented in the last sentence, beginning on line 19, is not convincing.

14. Section 3.3, paragraph 2: (a) The expectation of increased methanol concentrations in spring (lines 24-25) should be discussed later in this section, not in paragraph 2. (b) In sentence 3 (line 25), it is not the methanol emissions that are hampered by the dry spring, but rather the rate of plant growth. Are there specific rainfall data to support your statement in sentence 3? Even better, are there any measurements of net primary productivity in the region? (c) In sentence 4, beginning line 27, it is more likely that the methanol concentrations were greater in the summer (not because it was 'during the growing season) but because gas-phase concentrations of methanol were greater due to more rapid cell growth and/or enhanced temperatures. (d) The last sentence in this paragraph is difficult to understand, mainly because of the use of 'agrees'; if the argument presented in this sentence is important, explain it more clearly (even if it takes 2 or 3 sentences).

15. Section 3.3, paragraph 3: (a) It is not clear here how the dates for the growing and non-growing periods were obtained. Were they based on NPP measurements or other plant growth experiments? Without supporting evidence, the dates seem rather arbitrary. (b) That aspect to one side, you are not presenting a new argument here; biogenic emissions were already suggested as a major contributor to atmospheric methanol concentrations earlier in the paper. This paragraph, along with Fig. 5, could be omitted without any loss to the overall argument.

C45

16. Section 3.4, paragraph 1: (a) Sentence 3 (line 16): Given the frequent referral to biogenic and anthropogenic methanol in your paper, there is an argument for moving this section to a position earlier in the paper. On the other hand, unless one can estimate the relative percentages of biogenic and anthropogenic methanol, there is little value in distinguishing the two sources of methanol. (b) line 20: an  $r$  of 0.46 would not normally be considered as a strong correlation. (c) The last sentence of this paragraph is rather convoluted, but I gather it is saying that the methanol in the rain water has a primarily biogenic source. I don't however consider that the correlations in Table 1 provide strong proof for that assumption. If this sentence is retained in the paper, it must be rewritten.

17: Section 3.4, paragraph 2: (a) Some support for the theory that methanol is replaced via transpiration might come through a comparison of day- and night-time samples (assuming transpiration is much lower at night). (b) Because formaldehyde does not exhibit washout is not evidence for assuming that methanol will, or will not, exhibit washout (lines 3-5). This sentence is best deleted. (c) The last sentence in this section, which seems to follow from the preceding sentence, makes an assumption about methanol production mechanisms that are unsupported by evidence and are not included in the global budgets for methanol referred to in this study. However, if there is evidence in the literature that methanol is formed by photochemical reactions in rain-water, describe the reactions, and include the reference at the end of this statement. If not, omit the sentence.

18. Section 4: The material in this section is a mixture of implications, discussion, conclusions, and observations from other studies. There may be good reasons for taking this approach, but I would prefer to have a more traditional Conclusion section, with the other relevant material incorporated in the Discussion.

19: Section 4, paragraph 1: Some of the statement included in this paragraph have been commented on earlier in this review e.g. paragraph 1, sentence 3 (lines 12-14), sentence 5 (lines 16-18), and sentence 6 (lines 18-20). Consideration should be given

C46

to these earlier comments.

20. Section 4, paragraph 2: (a) Include uncertainties with the percentages given in this paragraph. (b) It is not clear why sentence 2 is included, particularly when the information is from another study. If you wish to compare the methanol content of the rainwater samples with formaldehyde and acetaldehyde, use measurements from this study, which presumably are available. But unless, you have corresponding gas-phase measurements, there doesn't seem to be any purpose in making these comparisons. (c) Re sentence 3, since you have measurements of methanol and DOC in each sample, the uncertainty in the 1.5% average should indicate the variability, without relying on 'the lack of correlation with DOC'

21. Section 4, paragraph 3: (a) The first sentence in this paragraph is relevant to this study, but it doesn't in itself indicate that wet deposition is a significant source in marine waters, since there are many other factors that control the concentration of methanol in the ocean. Revise the sentence. (b) It may be of value to discuss some of the other sentences in this paragraph as a lead-up to paragraph 4; but, as a reader of your paper, I would like to see the emphasis placed firmly on 'methanol concentrations in rainwater'. Unless, you have made methanol measurement in the ocean or in freshwater bodies in this study, the material following sentence 1 has little relevance to the theme of this study.

22. Section 4, paragraph 4: (a) I'm not sure that modellers would accept the results of this study 'as an appropriate proxy for global rainwater concentrations', but they might consider it as a first approximation for the mid-latitudes of the Northern Hemisphere when estimating a global wet deposition sink. It would be best to reword this sentence along this line. (b) Include an uncertainty for your estimate of 20 Tg yr<sup>-1</sup>, which includes the uncertainty in your measured concentration and also the uncertainty in global annual precipitation. (c) One would expect your estimate for a wet deposition sink to be significantly higher because it is relevant only for the location in which it was made. The gas-phase concentration of methanol is much less in the Southern Hemisphere

C47

(and also in the Arctic), as is the average annual precipitation. With that in mind, it wouldn't be too difficult to use your methanol concentration to estimate a reasonable global value. (d) The sentence in lines 19-22 is a little confusing as Tie et al. give only a range with no estimate of the mean; it would be better breaking this sentence into 2 sentences, the first of which compares your estimate to the theoretical range, and the second comparing your value to the global averages. (e) The sentence beginning on line 22 is a bit harsh; your wet deposition sink is much higher because you haven't estimated a wet deposition rate appropriate for the Southern Hemisphere (which is likely to be about half of that in the Northern Hemisphere). Nonetheless, I agree completely with the final sentence of the paragraph.

#### Technical Comments

Page 1376

line 4: insert a hyphen between 'volume' and 'weighted'. Check that this is done consistently throughout the paper.

line 7: insert 'by other authors' after 'waters'

line 7: insert a comma after 'by other authors'; and replace 'potential' by 'potentially'

line 7: insert 'of methanol' after 'source'

line 8: insert 'that' after 'Assuming'

line 9: insert a comma after 'concentrations'

line 9: insert a comma after 'yr<sup>-1</sup>', and insert 'that' after 'implies'

line 11: insert 'in rainwater' after 'concentrations', and insert 'significantly' after 'correlate'

line 12: replace 'suggest' by 'suggests'

line 14: insert 'The' before 'methanol'

C48

line 15: replace '+' by '±'

line 16: replace '+' by '±'

line 18: replace 'between' by 'during'

line 19: insert a comma after 'production'

line 21: the adjective 'higher' is used to indicate something that is at a greater elevation. It is better to use 'larger' or 'greater' in the context of a concentration. Check this throughout your paper.

line 21: insert a comma after 'origin'

line 22: replace 'origin' by 'origins'

line 25: insert a comma after 'troposphere'

line 26: If the one electron-dot is included in the hydroxyl radical symbol, it should be done consistently throughout the paper. Its inclusion (or exclusion) may be considered optional.

Page 1377

lines 1-2: insert 'a' before '1-2%' and before '1-3%'

line 5: including 'radicals' along with 'OH' is redundant

line 5: omit or replace 'primary'. Formic acid is produced by oxidation of formaldehyde following hydrogen abstraction of methanol, and thus the occurrence of formic acid is secondary production.

line 6: insert a comma after '2000)'

line 7: 'uptake' is a noun, and thus should be replaced by an appropriate verb, perhaps 'adsorb'

line 9: insert 'assessing' after 'in'

C49

line 10: replace 'lead' by 'led'

line 14: 'biomass burning' could be considered biogenic as it does not require the intervention of humans.

line 14: not clear to what 'gasoline additives' you are referring, other than methanol itself. If so, 'evaporation of methanol-based fuels' might be better.

line 14: omit 'other'

line 15: The citation for 'Howard, 1990' is not included in your References. Also, it is preferable to use primary sources of this information i.e. studies that have identified these sources (see Wells et al. 2012 for examples). Alternatively, you might say 'as summarized by Wells et al. 2012'

line 15: Note that the word 'while' is used to connect two clauses that are coincident in time e.g. "While I am reading, I am making notes". In this case, replace 'While' by 'Whereas'

lines 15-16: replace 'that' by 'that, globally,'

line 17: replace 'debate' by 'investigation'

line 19: replace 'while' by 'whereas' or 'but'

line 20: replace 'This' by 'The'

lines 20-23: This sentence is not necessary; but, if you wish to include it, I suggest replacing 'which are represented by higher methanol ...' by "that have higher methanol ...", and add 'than rural areas' at the end of the sentence.

line 23: replace 'Jacob' by 'Jacob et al.'

line 23-25: This sentence could be confusing. It depends what 'regions' you are considering, and how you intend to 'differentiate'. The sentence needs to be revised in order for a reader to understand the point you are trying to make.

C50

line 26: omit 'Earlier'

p. 1378

line 1: insert commas before and after 'in turn'

line 2: omit 'and sink'. Replace 'and sink' by 'and the sink from'. Omit 'respectively.

line 3: For multiple references, list in chronological order.

line 5: replace 'its' by 'their'

line 7-8: 'Tg yr<sup>-1</sup>' is a measure of emission rate, not flux.

line 15: replace 'is' by 'are'

line 16: insert a hyphen in 'aqueous-phase'

line 19: omit 'on an event basis', as it is also stated in line 21.

line 26: better to use past tense in this sentence – replace 'can' by 'could'

p. 1379

line 1: better to terminate the sentence after 'collection', and begin the new sentence with 'This reduces the ..'

line 1: replace 'time' by 'times'

line 5: replace 'combusting' by 'heating'

line 7-8: The citation for 'Topal et al., 1985' is not included in your References.

line 9: delete 'together'

line 11: replace 'derivtization' by 'derivatization'

line 12: include acronym '(DNPH)' after '2,4-dinitrophenylhydrazine', as it is used later in line 25

C51

line 18: preferable to replace 'DIW' by 'deionized water'  
line 20: replace 'on' by 'from'  
line 24: delete comma after 'buffer' and replace with 'along with'  
line 28: replace 'RSD' by 'relative standard deviation'

p. 1380

line 9: insert 'the' after 'in', and also after 'of'  
line 14: replace 'was' by 'were'  
line 17: insert 'the' after 'in'  
line 22: replace 'Organic carbon' by 'The dissolved organic carbon'  
line 22: insert 'the' after 'in', and replace 'were' by 'was'  
line 24: 'Willey et al, 2000' is not included in your References

p. 1381

line 10: 'NOAA/ARL, 2013' is not included in your References. Also, be consistent with the punctuation in in-text references; see also line 28 on this page.

line 11: delete semicolon after 'model'

lines 13-14: it is not necessary to include acronyms if they are not used later in the paper

line 17: it is not usual to include a hyphen in 'air mass', but if including the hyphen, be consistent throughout the paper; cf. line 20

lines 20-22: omit the final phrase 'and likewise over ocean for marine types' and either include a new sentence to define marine trajectories, or revise the sentence to read 'Terrestrial or marine air masses are those . . . .over a landmass or ocean, respectively'.

C52

line 24: insert 'were those that' after 'trajectories'  
line 26: replace 'size normalized county level' by 'size-normalized county-level'  
line 27: insert 'The' before 'methanol'  
line 28: Use upper case for 'Emissions Inventory'  
line 28: 'NEI, 2008' is not included in your References

p. 1382

line 2: insert 'in the samples' after 'concentrations'; and insert 'the' after 'for'  
line 5: Snider and Dawson's study was conducted in 1982.  
line 6: 'Snider and Dawson, 1985' is not cited in your References  
line 7: insert a comma after 'however'  
line 8: insert a comma after 'concentration'  
line 9: delete 'more' and insert 'variation' after 'temporal'  
line 17: replace 'higher' by 'larger' or 'greater'  
line 19: replace 'gas phase' by 'gas-phase'  
line 21: The article cited as 'Jacob, 2005' has a number of authors, and should be written as 'Jacob et al., 2005'. Be sure to correct this throughout the paper, and also in your References  
line 23: replace 'lower' by 'smaller'

line 24: 'significant' is not the right adjective here. A concentration is significant if it above the detection level. What needs to be considered is whether methanol concentrations in marine air-masses are significantly less or greater than terrestrial types.

line 25: insert "in air masses of marine origin" after 'amounts'

C53



line 25: replace the period after 'reasons' by a colon. Also replace 'can' by 'may'

line 26: insert a comma after 'sink'

p. 1383

line 1: use a semi-colon rather than a period after 'atmosphere'

lines 1-2: remove the parentheses around '5 to 12'

line 2: insert a comma after '(Jacob, 2005)'

line 2: replace 'oceans' by 'the ocean'. Also, replace the period by a semi-colon.

line 3: replace 'would' by 'could'

lines 3-4: since it is the methanol that would contain some terrestrial sources, reword the sentence as 'methanol at the rain collection site could include some methanol scavenged from local terrestrial sources'.

line 6: replace 'Methanol concentrations' by 'The methanol concentrations measured in this study'

line 6: delete 'further'; and replace 'subdivided' by 'divided'

line 7: replace 'concentration' with 'the concentrations'

line 7: insert a hyphen between 'short' and 'term', and terminate the sentence after '(Fig. 3).'

line 8: change 'others' to 'other'

line 10: replace 'volume weighted' by 'average volume-weighted'

line 12: insert a comma after '6 p.m.)'

lines 12-13: replace 'a volume weighted' by 'an average volume-weighted'

line 14: replace 'to' by 'with'

C54

line 16: insert a comma after '2005)'

lines 17-20: improve the structure of this sentence.

line 19: insert a hyphen between 'light' and 'stimulated'

lines 19-22: list references chronologically

line 21: replace 'brings' by 'produces'

line 22: include a comma after 'increase'

line 24: Replace 'A ~ 6 fold' by 'The approximately 6-fold'

line 24: insert 'in this study' after 'night'

lines 24-26: there may be other causes for a decrease in methanol concentration at night' It does not automatically 'imply' and increase in dry deposition. Also, indicate to what surface uptake you are referring e.g., soil absorption, plant metabolism, etc.

line 26: insert "in the aqueous-phase concentration of methanol" after 'decrease'

line 27: insert a hyphen between 'water' and 'soluble'

line 28: Start a new sentence after 'collection site'

p. 1384

line 4: replace 'The methanol concentration was' by 'The methanol concentration data collected in this study were'

line 4: insert 'seasonal' after 'time'

line 5: insert 'the' after 'in'; and replace 'Seasons' by 'The seasons'

line 9: replace 'stronger' by 'large'; insert a comma after 'source'; replace 'it' by 'gas-phase methanol concentrations'; and replace 'lower' by 'smaller'

line 10: insert 'when plants are dormant' after 'months'

C55

line 11: list references chronologically; and insert a comma after '2005'  
line 13: insert a comma after 'instance'  
line 15: insert 'that' after 'estimated'; and replace 'having' by 'had an'  
line 16: insert a comma after 'decay'  
line 17: insert 'of methanol' after 'source'; and replace 'low' by 'small'  
line 18: it is not clear to what 'plant growth total' refers.; is it NPP?  
lines 18-19: list references chronologically  
line 20: insert 'the' after 'that'  
line 21: insert a comma after '2008'; and omit 'in turn'  
line 21: The citation for 'NC Drought, 2008' is not included in your References  
line 22: insert 'otherwise' after 'would'  
line 23: replace 'volume weighted concentration of methanol' by 'average volume-weighted concentrations of the methanol observed in this study'  
line 24: replace 'are' by 'were'  
line 26: insert a comma after 'seen'  
line 28: insert in 'rain water' after 'concentrations'; replace 'highest' by 'greatest'; and insert a comma after 'summer'  
line 29: better to say 'the plant growing season occurs during the summer' instead of 'summer is during growing season'  
line 29: insert a comma after 'season'  
p. 1385

C56

line 2: insert 'the' after 'in'; and replace 'concentration' by 'concentrations'  
lines 2-5: this sentence needs re-writing to improve clarity – perhaps expanding it into 2 or 3 sentences  
line 8: replace 'Methanol volume weighted concentration' by 'The average volume-weighted concentrations of methanol'  
line 9: replace 'that of' by 'those measured during'  
line 10: terminate the sentence after '0.1  $\mu\text{M}$ '; and construct another sentence using the remainder of the information  
line 11: 'fluctuation' is not the right choice of word here.  
line 15: replace 'correlation with each other' by 'intercorrelation'  
line 16: replace 'suggest' by 'suggests'  
line 17: replace 'alcohol' by 'methanol'  
line 18: insert '(DOC)' after 'carbon'  
line 19: insert 'that' after 'indicating'; replace 'the alcohol' by 'methanol'; and replace 'make' by 'makes'  
line 21: insert 'that' after 'suggesting'  
line 22: insert 'is' after 'methanol'; and insert 'of methanol' after 'concentration'  
line 24: insert 'significant' after 'no'; (Table 1 would be a better references than Fig. 6)  
line 25: delete 'simply'; and insert a comma after 'location'  
p. 1386  
line 7: Fig. 6 does not support this statement  
line 11: insert 'that' after 'suggests'

C57

line 15: replace 'higher' by 'larger'

line 21: insert a comma after 'content'; and insert 'that' after 'indicating'

p. 1387

line 1: insert 'samples measured in this study' after 'rainwater'

line 2: replace '(Dixon et al., 2011)' by 'by Dixon et al. (2011)'

line 2: insert 'of methanol' after 'deposition'; and replace 'potential' by 'potentially'

line 4: replace 'of' by 'on'

line 6: include reference at the end of this sentence

line 7: insert a comma after 'significant'

line 8: insert a comma after 'dominant'; and replace 'to' by 'in'

line 9: insert 'that' after 'likely'

line 10: insert a comma after 'significant'

line 11: delete the apostrophe in 'month's', and insert a comma after 'months'

line 13: include a reference at the end of this sentence.

line 14: delete 'most'; and replace 'uncertainties' by 'issues'

line 15: insert 'and uncertainty' after 'magnitude'; and replace 'flux' by 'rate'

line 16: replace 'rain' by 'sampling'

line 17: insert a comma after 'concentrations'

line 18: 'Pidwirny 2008' is not included in your References. Insert a comma after 'Pidwirny'

line 18: replace 'global methanol rainwater concentrations' by 'a global average rain-

C58

water concentration of methanol'

line 23: insert 'that' after 'suggest'

line 25: replace 'earth' by 'Earth'

line 26: 'labile' means 'subject to change'; this doesn't seem to apply in this context.

Table 1: in title, replace 'between' by 'among'

Figure 1 caption: insert 'Average' before 'volume-weighted' and change 'concentration' to 'concentrations'. Also, indicate that the error bars represent  $\pm$  one standard deviation from the average.

Figure 2 caption: replace 'county level' by 'county-level'

Figure 3: include a more descriptive caption. Include the last sentence in the first paragraph of Section 3.2. Also, indicate what the error bars represent.

Figure 4: include the number of samples in each season. The caption does not seem to represent what has been plotted. It seems that what is plotted is the 'average of the volume-weighted methanol concentrations in each season'. Indicate what the error bars represent.

Figure 5: insert a hyphen in 'non growing'. Revise the caption as suggested in the comments for Figures 1, 3 and 4.

Figure 6: replace 'rain fall' by 'rainfall' in both the x-axis title and caption.

---

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 1375, 2014.

C59