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Interactive Comment

Interactive comment on "A comparison of four receptor models used to quantify the boreal wildfire smoke contribution to surface PM_{2.5} in Halifax, Nova Scotia during the BORTAS-B experiment" by M. D. Gibson et al.

M. D. Gibson et al.

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Received and published: 25 November 2014

Author Response (AR): Firstly, thank you very much for taking the time to review the manuscript and providing excellent comments and suggestions to improve the manuscript!

Reviewer comment (RC): Table 7 is not required. The RMSE values from this table can be added to the respective Figures 5, 6, 7 and 8. The n, R2 and bias values from this table are already on the relevant figures and the mean obs and mean predicted data

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do not provide useful insight – the nature of the scatter plots in the figures provide the informative insight into model performance.

AR: Indeed, as you point out it is really not required. I have removed Table 7 and blended the RMSE detail into the Figures 5, 6, 7 and 8 as requested. I have also added the C.I. (95%) of the slope and intercept to the Figures.

P24058, I25. I have removed the sentence 'Table 7 summarizes the four receptor model parameters used for predicting PM2.5 during the BORTAS-B experiment' as it is now redundant.

P24058, I26. Because of the above change I altered 'Table 8' to 'Table 7' to now read 'Table 7 presents the woodsmoke source apportionment descriptive statistics for each receptor model.'

RC: P24045, I25: It is not clear what is meant by the statement that burning forests is a significant source of secondary trace gases. By definition, secondary species are not emitted. Is it meant that burning forests is a significant source of precursor gases for formation of secondary components of PM? Rephrase as appropriate.

and

RC: P24045, I26: The phrasing that forest fires are a source of 'size-resolved particulate matter' also does not make scientific sense. Rephrase along the lines of ": : :.and particulate matter (PM) of different size fractions to the: : :"

AR: Thank you, very good advice. P24045, I26: I have changed the sentence to simply read 'The burning of these forests is a significant source of gases and airborne particulate matter (PM) of different size fractions (Drysdale, 2008).

RC: P24046, I17: Change "will explore" to "explores".

AR: P24046, I17: I have changed "will explore" to "explores".

RC: P24046, I20: Rephrase the start of the sentence more directly as: "A number of

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different receptor modelling approaches are utilized..."

AR: P24046, I20: I have changed the sentence to read 'A number of different receptor modelling approaches are utilized for the source apportionment of PM2.5, e.g. multivariate least squares factor analysis approaches such as Positive Matrix Factorization (PMF), Pragmatic Mass Closure (PMC) methods and Chemical Mass Balance (CMB) source profile techniques (Gibson et al., 2013b, 2009; Ward et al., 2004; Gugamsetty et al., 2012; Harrison et al., 2011).

RC: P24048, I17 & I18: Rephrase to avoid repetition of "quantitative comparison"

AR: I have changed the sentence on P24048, L17 to read 'The objective is to determine the ability of these models to predict overall PM2.5 mass and the contributions of minor components.'

RC: P24048, I27: Replace "DGS" here with "Dalhousie Ground Station (DGS)", as this is the first point in this section when DGS is used.

AR: P24048, I27. As requested, I have replaced DGS with "Dalhousie Ground Station (DGS)"

RC: P24049, I2: Replace "Dalhousie Ground Station (DGS)" with "DGS".

AR: P24049, I2: I have replaced "Dalhousie Ground Station (DGS)" with "DGS".

RC: P24048, I5: < Should be P24049, I4. Delete the sentence beginning "The PM2.5 mass..."; this sentence restates what the reader already knows about this paper.

AR: P24049, I4: I have deleted the sentence 'The PM2.5 mass and chemical components were used in the four receptor models presented here.'

RC: P24049, l8: delete the word "ion" (It is superfluous as the chemical formula shows it is an ion; also the word ion is not used after nitrate).

AR: P24049, I8: I have deleted the word 'ion' after 'ammonium'.

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RC: P24049, I9: delete "ion".

AR: P24049, I9: I have deleted the word 'ion' after 'sulfate'.

RC: P24051, I9: reverse the words "PM2.5" and "the" to make grammatical sense.

AR: P24051, I9: I have reversed the words "PM2.5" and "the".

RC: P24051, I11: The sentence beginning "With the GEOS-5: : :." does not make grammatical sense. Rephrase.

AR: P24051, I11: I have rephrased the sentence to read 'The GEOS-5 forecast model also provided additional evidence that upwind wildfire PM2.5 impacted the surface in Halifax during the BORTAS-B experiment.

RC: P24051, I15-I17: The two sentences of this opening paragraph can be deleted as simply repeating what the reader already knows about the work in this paper.

AR: P24051, I15-I17: I have deleted the two sentences 'In this study, we compare the results of four receptor models for estimate the source of PM2.5 aerosol to Halifax during the BORTAS-B campaign. We describe these four models here.'

RC: P24051, I20-I23: The two sentences starting "Leveglocosan was added...." and "The addition of levoglucosan..." repeat almost the same thing. Replace with a single sentence.

AR: P24051, I20-I23: I have replaced the two sentences with 'In this manuscript, levoglucosan was added to the previously modelled PM2.5 speciated data as a means to unambiguously identify the presence of woodsmoke (Gibson et al. 2013b)'.

RC: P24052, I7: Replace "included" with "were".

AR: P24052, I7: I have replaced "included" with "were" so that the sentence will read 'In the previous manuscript by Gibson et al. (2013b), six major sources were identified and were Long-Range Transport (LRT)'

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RC: P24054, I19: Add the unit after the bias value 1.3.

AR: P24054, I19: The sentence now reads '..... bias of 1.3 μ g m-3.'

RC: P24054, I23: The sentence starting "From Fig. 3: : " does not make grammatical sense. Rephrase.

AR: P24054, I23: I have changed the sentence 'From Fig. 3 it can be seen that the CMB intercept was located at $-0.53~\mu g$ m-3, a slope of 1.0, R2 of 0.88 and a bias of 4.3.... to read 'It can be seen in Fig. 3 that the CMB model intercept was located at $-0.53~\mu g$ m-3, it has a slope of 1.0, an R2 of 0.88 and a bias of 4.3 μg m-3.'

RC: P24054, I25: Add the unit after the bias value 4.3.

AR: P24054, I25: I have added the unit after the bias value 4.3 so that the sentence reads '...CMB model intercept was located at $-0.53~\mu g$ m-3, it has a slope of 1.0, an R2 of 0.88 and a bias of 4.3 μg m-3.'

RC: P24054, I28: Insert "for" before "CMB".

AR: P24054, I28: I have inserted "for" before "CMB" so that the sentence reads 'While the PMF bias is better than for CMB, '

RC: P24055, I3: I don't think "parsimonious" is the appropriate word here. Parsimonious indicates most restricted or most efficient. Perhaps most "useful" is more appropriate.

AR: P24055, I3: Thank you. I have changed the sentence to read 'However, because PMF predicts the PM2.5 mass on all sample days, has a slope of 0.88 and the ability to predict very low PM2.5 mass concentrations, often seen in Halifax, in these respects it is the most useful of the four receptor models.'

RC: P24055, I9: The start of this sentence can be written more directly as "Tables 3-6 show That..."

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AR: P24055, I9: I have changed the sentence to read 'Tables 3–6 show that the four receptor models identify different number and type of PM2.5 source respectively, e.g. '

RC:P24055, I10: < should be P24056, I10: The word "co-vary" is probably better hyphenated, otherwise it looks odd.

AR:P24055, I10: < should be P24056, I10: I have changed the sentence to read 'Conversely, PMF and APCS have other mass contributions that co-vary with the LRT....'

RC: P24056, I14: The sentence starting "Because..." is too long. Suggest starting a new sentence after "source" with "Instead, the LRT..."

AR: P24056, I14: I have split the one sentence into two as suggested to now read. 'Because of co-varying species associated with the LRT NH4NO3 in the APCS and PMF models, NH4NO3 could not be factored into a pure apportioned source. Instead, the LRT NH4NO3 in both APCS and PMF is also associated with other LRT species, e.g. OM, BC, Na and is referred to as LRT Marine Mixed PM2.5 as the NH4NO3 was likely mixed with aged marine aerosol as the air mass crossed the Gulf of Maine and the Bay of Fundy en route to Halifax.

RC: P24056, I23: Replace "related to" with "from".

AR: P24056, I23: I have replaced "related to" with "from".

RC: P24057, I4: The sentence beginning "To identify..." is both too long and doesn't make grammatical sense. Rewrite.

AR: P24057, I4: I have re-written the sentence to read 'To help identify upwind forest fire source regions, we used a combination of visible MODIS satellite images, MODIS fire hot spot maps, 5 day HYSPLIT air mass back trajectories (Gibson et al., 2013b), FLEXPART air mass trajectories (Stohl et al., 2005) chemical transport models (Palmer et al., 2013), Raman Lidar (Bitar et al., 2010) and aircraft measurements (Palmer et al., 2013). Together, these approaches helped corroborate the woodsmoke event impacting Halifax on 21 July.

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RC: P24057, I12: The "(c)" should come after "DGS".

AR: P24057, I12: The "(c)" has been moved to come after "DGS".

RC: P24057, I20: Replace the word "parsimonious" with a more appropriate word.

AR: P24057, I20: I have replaced the word "parsimonious" with 'useful'.

RC: P24057, I22: Should read "contained in"?

AR: P24057, I22: I have changed 'contain in' to read "contained in".

RC: P24057, I28: USA is the name of the country, so better to replace "from the NE

US" with "from NE USA".

AR: P24057, I28: I have changed "from the NE US" to read "from NE USA".

RC: P24058, I2: Replace "from the NE US" with "from NE USA".

AR: P24058, I2: I have changed "from the NE US" to read "from NE USA".

RC: P24058, I4: Replace "from the NE US" with "from NE USA".

AR: P24058, I4: I have changed "from the NE US" to read "from NE USA".

RC: P24058, I11: Two sentence here can be joined and many words cut: "......come from the aircraft column profiles for CO, acetonitrile and aerosol backscatters shown in Figure 10d."

AR: P24058, I11: I have combined the two sentences to now read 'Figure 10d shows aircraft column profiles for CO, acetonitrile and aerosol backscatter, which provide further forensic evidence of woodsmoke impacting the DGS in Halifax.'

RC: P24058, I25: See comment above, delete Table 7 and reference to it, and add (and cite to) statistical data to Figures 5-8 as appropriate.

AR: P24058, I25: I have deleted Table 7 and placed the RMSE values in Figures 1-4 as requested earlier. I have removed the sentence 'Table 7 summarizes the four receptor

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model parameters used for predicting PM2.5 during the BORTAS-B experiment.' The new sentence reads 'Details of the performance parameters related to the four receptor models are provided in Figure 1 - 4.'

RC: P24059, I9: < should be I8. Insert "and its" before "known".

AR: P24059, I6: As requested, I have inserted "and its" before "known". The sentence now reads 'However, because of the PMF model's better PM2.5 predictive capability (especially below 2.0 μ g m-3) and clear woodsmoke marker source identification, and its known statistical robustness over APCS, its results are likely the most accurate of the four models.

RC: P24059, I27: Delete "relative" (the sentence presents absolute PM2.5 contributions, not relative contributions).

AR: P24059, I27: I have deleted "relative" from the sentence to now read 'The median (min: max) woodsmoke contribution to PM2.5 estimated using PMF was found to be 0.14....'

Note to Editor and Print Office Here is the full reference for Wheeler et al. (2014) that is now in print.

Wheeler, A. J., Gibson, M. D., Macneill, M., Ward, T. J., Wallace, L. A., Kuchta, J., Seaboyer, M., and Dabek-zlotorzynska, E.: Impacts of air cleaners on indoor air quality in residences impacted by wood smoke, Environ. Sci. Technol., 48, 12157-12163, 2014.

I have also created a new figure that we feel is a valuable addition to the paper as shown below.

This new figure would be Fig 5 (attached as New Fig 5). New Fig 5. Title would be NEW Figure 5. PMF chemical species and their factor source profiles contributions

Therefore Figure 6, 7, 8, 9, 10, 11 and 12 would become Figures 7, 8, 9, 10, 11, 12

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and 13 respectively.

P24052, I12. The sentence 'The PMF factor profile used to identify woodsmoke contained 99% of the levoglucosan mass.' Would be removed.

P24055, I5, new sentences added 'Figure 5 provides the PMF model output associated with the chemical species and their factor source profiles contributions. It can be seen in Fig 5. that the PMF factor profile used to identify woodsmoke contained 90% of the total levoglucosan mass.

Therefore, the following minor changes would need to be made to the manuscript incorporate the new figure.

P24055, I5, 'Figures 5-8 provide ...' would be changed to 'Figures 6-9 provide ...'

P24055, I7, '... with Figs. 5-8 are ...' changed to '...with Figs. 6-9 are ...'

P24055, I27, '... reference to Fig. 6,' changed to '...reference to Fig. 7,"

P24056, I22 '....series plot shown in Fig. 9.' Changed to '....series plot shown in Fig. 10.'

P24056, I22 '....feature of Fig. 9....' Changed to '....feature of Fig. 10 is'

P24056, I28 '....seen from Fig. 9 that' Changed to '....seen from Fig. 10 that'

P24057, I9. 'Figure 10 provides ...' changed to 'Figure 11 provides ...'.

P24057, I15. 'Figure 10a shows ...' changed to 'Figure 11a shows ...'.

P24057, I19. '... in Fig. 10a are ...' changed to '.... In Fig. 11a are ...'.

P24057, I20. 'Fig5' changed to 'Fig 6'.

P24057, I22. '...in Fig. 10.' changed to '... in Fig 11.'

P24057, I23. '...in Fig. 8.' changed to '... in Fig 9, it can ...'

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P24058, I2. '...from Fig. 10b that...' changed to '... from Fig. 11b that ...'
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P24058, I3. '...in Fig. 10 a.' changed to '... in Fig. 11a.'

P24058, I6. '... timeseries (Fig. 8) was ...' changed to '...timeseries (Fig. 9) was...'

P24058, I8. '...from Fig. 10c that...' changed to '...from Fig. 10c that...'

P24058, I11. '...in Fig. 10d.' changed to '...in Fig. 10d.'

P24058, I11. 'Fig. 10d shows ...' changed to 'Fig. 11d shows'

P24058, I13. '... in Fig. 10d points ... 'changed to '... in Fig. 11d points ... '

P24058, I14. 'Figure. 11 provides' changed to 'Figure 12 provides....'

P24058, I17. '... shown in Fig. 9 and 10.' changed to '... shown in Fig. 10 and 11.'

P24058, I18. '... shown in Fig. 9 on ...' changed to '... shown in Fig. 10 on ...'

P24058, I19. '... seen in Fig. 12 where ...' changed to '... seen in Fig. 13 where ...'

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 24043, 2014.

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6 8 10 12 14 16 18 Observed PM_{2.5} [µg m⁻³] Absolute Principal Component Scores (APCS) predicted vs. observed PM_{2.5} Predicted PM_{2.5} [µg m⁻³] Observed PM_{2.5} [µg m⁻³] b) Pragmatic Mass Closure (PMC) predicted vs. observed PM_{2.5} Predicted PM_{2.5} [µg m·3] ---- 1:1 Line 6 8 10 12 14 16 18 Observed PM_{2.5} [µg m⁻³] c) Chemical Mass Balance (CMB) predicted vs. observed PM_{2.5} Predicted PM_{2.5} [µg m³] 10 12 14 16 18 Observed PM_{2.5} [µg m⁻³] d) Positive Matrix Factorization (PMF) predicted vs. observed PM_{2.5}

Fig. 1. Four receptor model comparison predicted vs. observed panel

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LRT Pollution (NH₄)₂SO₄ LRT Pollution Coal/Industry T × N T Z z z > V D .. B Q X z u g Ship Emissions Species

Fig. 2. PMF source factor profiles

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■LRT Pollution (NH4)2SO4 LRT Pollution Aged Marine Aerosol --Observed PM2.5 a) Time series of the relevant source contributions to PM2 s estimated by Absolute Principal Component Scores (APCS) 18.0 17.0 16.0 15.0 14.0 13.0 12.0 5 10.0 5 9.0 7.0 6.0 5.0 4.0 3.0 2.0 1.0 Particle Bound Water Sea Salt Trace Element Oxides Surface Dust Black Carbon Organic Matter LRT Pollution (NH4)2SO4 LRT Pollution NH4NO3 b) Time series of the relevant source contributions to PM_{2.5} estimated by Pragmatic Mass Closure (PMC) receptor model time series 18.0 17.0 16.0 15.0 12.0 12.0 (c) 11.0 (c) 9.0 9.0 6.0 6.0 4.0 2.0 1.0 0.0 0.0 c) Time series of the relevant source contributions to PM_{2.5} estimated by Chemical Mass balance (CMB) d) Time series of the relevant source contributions to PM_{2,5} estimated by Positive Matrix Factorization (PMF)

Fig. 3. Four receptor model source apportionment panel

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16.0 5.0 4.5 Woodsmoke-PM_{2.5} [µg m⁻³] 14.0 4.0 12.0 3.5 10.0 3.0 2.5 8.0 2.0 6.0 1.5 4.0 1.0 2.0 0.5 0.0 27.07.22 19.07.22 22.07.22 25.07.12 21.07.22 29:07:12 31.07.11 22.08.22 1301.11 1501.11

•CMB

PM2.5

Fig. 4. Four model woodsmoke time series plot

-- APCS

· · · PMC

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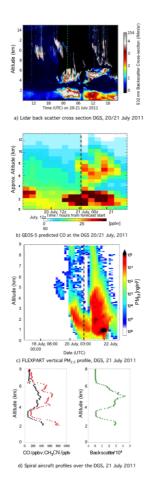


Fig. 5. Lidar_GEOS 5_FLEXPART_Aircraft Spiral panel

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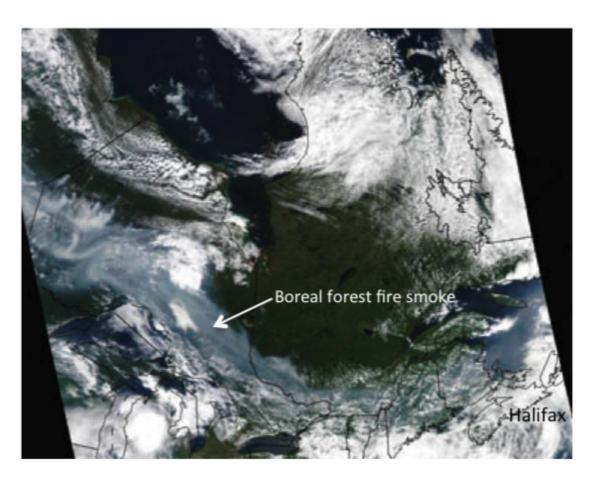


Fig. 6. MODIS image of smoke plume

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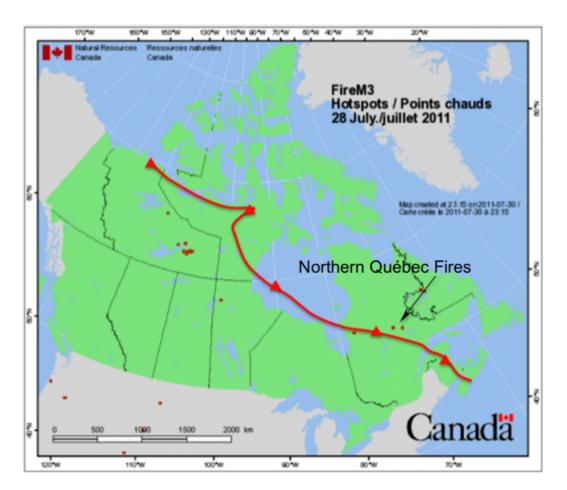


Fig. 7. MODIS fire hot spot map and back trajectory

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