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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Identifying the sources driving observed $PM_{2.5}$ variability over Halifax, Nova Scotia, during BORTAS-B" *by* M. D. Gibson et al.

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Anonymous Referee #3

General comments The paper entitled "Identifying the sources driving observed PM2.5 variability over Halifax, Nova Scotia, during BORTAS-B" by Gibson et al., discusses PM2.5 concentrations and chemical composition recorded over Halifax (Nova Scotia) from 11 July to 26 August 2011. The campaign was performed in the framework of the B Phase of an experiment directed towards the quantiïňĄcation of the impact of BOReal forest ïňĄres on Tropospheric oxidants over the Atlantic using Aircraft and

Satellites (BORTAS). The United States Environmental Protection Agency (US EPA) Positive Matrix Factorization (PMF) receptor model was used to determine the average mass as well as percentage source contributions to the PM2.5 measured during the sampling campaign. The factor identiïňĄcation was based on chemical markers typical of speciïňĄc sources and was supported by air mass back-trajectory analysis and local wind direction.

I read the manuscript in depth and I am afraid that no real advance can be found in this study, in addition the work highlights no original or new ïňĄndings. In fact, the methodology applied is not so new, and similar studies have already been carried out. However, the major remark is that the interpretation of the results appears quite weak. To my mind, much effort has been made to explain both of the instrumentation technical details and the well-known methodologies applied rather than discuss the results obtained. Moreover, the study, as it is presented, seems to be of local rather than regional or global interest. To end with, I think that the entire discussion needs to be improved and better contextualized in order to be made suitable for publication on the Atmospheric Chemistry and Physics, particularly on a special issue.

Authors Response Firstly, thank you for taking the time to review the paper and for your very useful and insightful comments that we feel has greatly improved the paper.

However, we have an alternative point of view regarding your conclusion on the appropriateness of the paper for publication in ACP and that this study offers nothing new or of international interest.

This study was designed to be a critical component of the ground-based measurements made during the BORTAS-B campaign. The aim of providing PM2.5 chemical speciation data during BORTAS-B was to a) verify the PM2.5 forecast runs that helped to guide the aircraft into the wildfire plumes advecting over Halifax and impacting the surface b) to be used for comparison with the other surface gas and size-resolved particle number measurements (described in Palmer et al 2013) for a forth coming ground

station measurement intercomparison paper and c) to understand the upwind and local drivers of the PM2.5 temporal variability over Halifax during BORTAS-B and not just wildfire smoke contributions to PM2.5. The PM2.5 instruments were co-located with other instruments at the Dalhousie Ground Station that included a Lidar, Sun Photometer, two atmospheric column FTIR's, particle counters, O3, SO2 (Palmer et al 2013.). This paper provides valuable insights into the PM2.5 chemistry at the surface and supports both the atmospheric modelling, other ground based measurements and aircraft measurements taken over Halifax during BORTAS-B. Therefore, the BORTAS-B researchers and our peers see this paper as a vital addition to the BORTAS-B special issue of ACP.

Another reason this paper is of importance and worthy of publication in ACP is because to the vastly improved temporal resolution of the PM2.5 chemical speciation data. The Federal Government PM2.5 instruments were not functioning in Halifax during BORTAS-B. We were therefore faced with collecting our own Federal Equivalent Method data that covered the BORTAS-B campaign. Our observations were made at a much greater temporal resolution than would normally be taken by the Government (every 24-hrs versus every 6th day). The collocation of continuous black carbon and organic matter is new to the Atlantic region of Canada and provides a new and unique data set that, together with the daily Federal Reference Method data we collected IS definitely of international interest.

SpeciïňĄc comments 1.Introduction The introduction does not ïňĄt well with the aim of the study.

Authors Response The abstract has had the additional text added: "This paper presents the results of the PMF receptor modelling, providing valuable insight into the local and upwind sources impacting surface PM2.5 in Halifax and a vital data set for comparison with other collocated ground based observations of atmospheric composition made during BORTAS-B".

The introduction has been re-written as follows:- Numerous studies have shown an association between exposure to ambient fine atmospheric particles, less than or equal to a median aerodynamic diameter of 2.5 microns (PM2.5), and acute and chronic health effects (Pope et al., 2002;Dominici et al., 2006). Studies have shown that biomass derived PM2.5 is at least as harmful to health as fossil fuel combustion related PM2.5 (Allen et al., 2008;Norris et al., 2000). In addition, forest fire derived PM2.5 chemical components and associated gases are known to impact climate and local air quality (Parrington et al., 2011;Gambaro et al., 2008). Remote sensing estimates of the 10-yr average number of forest fires each year in North America is 5,062, covering an area of 1,323,736 ha, making biomass burning a major source of PM2.5 in North America (Palmer et al., 2013). Because of the importance of understanding the impact of North American boreal forest wildfires on northern hemisphere tropospheric chemistry, a multi-national project, led by the University of Edinburgh, was conducted out of Halifax, Nova Scotia, Canada during the summer of 2011. The study aim was to quantify the impact of "BOReal forest fires on Tropospheric oxidants over the Atlantic using Aircraft and Satellites". Central to BORTAS-B was a measurement campaign with the UK Facility for Airborne Atmospheric Measurements (FAAM) BAe146 research aircraft (Parrington et al., 2011;Palmer et al., 2013). In addition, numerous satellite observations of trace pyrogenic gases were made (Tereszchuk et al., 2012). One important component of the BORTAS-B project was the Dalhousie University Ground Station (DGS) in Halifax. The DGS was established to determine the temporal variability of size-resolved particulate composition and gas species concentrations both in-situ and through the atmospheric column. These measurements were used to help validate air quality forecast models used to guide the BA3146 aircraft toward wild fire plume outflows from within and exiting Eastern Canada, to validate satellite surface and column composition observations over Halifax, to validate Lidar surface and column observations over Halifax, for identifying wild fire smoke plumes as they passed over or impacted the surface in Halifax and used for additional insight into the atmospheric chemistry prevalent during the BORTAS-B campaign. This paper presents the chem-

ical speciation and mass concentration of atmospheric fine particulate matter composition less than, or equal to, a median aerodynamic diameter of 2.5 microns (PM2.5). Receptor modelling of the PM2.5 mass and chemical species was used in this paper to identify the local and upwind sources responsible for driving the observed temporal variability of PM2.5 in Halifax sampled during the BORTAS-B mission.

Change in conclusion:- The PMF model was used to determine six major sources contributing to the PM2.5 mass sampled during the BORTAS-B study. Although other BORTAS-B related observations (Palmer, 2012) showed that transient Boreal wildfire smoke plumes did pass over and impact the surface in Nova Scotia, there was insufficient mass for PMF to apportion wildfire smoke to the PM2.5 observed in Halifax. It was shown that the dominant source contribution to summertime PM2.5 mass in Halifax was from LRT Pollution with a contribution from aged marine aerosol (75%) coincident with SW air flow. This is consistent with the conventional wisdom that Nova Scotia is the "tail pipe of North America". Comparison of the PMF total PM2.5 mass with the observed total PM2.5 mass over the sampling period showed good agreement (R2 = 0.87, bias = 0.17 and RSME = 1.5 μg/m3), demonstrating the PMF receptor model performed well. The individual PM2.5 species and source apportionment data provides valuable comparative information that can be used to interpret other collocated ground based measurements of atmospheric composition made at the Dalhousie Ground Station during BORTAS-B. In addition, this study provided valuable new insight into the major local and upwind sources driving the temporal variability of surface PM2.5 at the DGS during BORTAS-B. The study highlights the utility of using air mass back trajectories coupled with local wind direction dependence to help identify the source of PM2.5. The techniques used in this study show considerable promise for further application to other sites and to identify other source categories of PM2.5.

This study provides vital new data that will be useful for intercomparison with other the DGS BORTAS-B observations that will follow. investigations as well as new information that can be used for population air pollution exposure assessment, air quality

management and urban planning in Halifax.

Please revise it. 2.Measurements General comment: Excessive technical details were provided. This section needs a deep revision. In particular, it should be considerably shortened. Page 4495 lines 8-10: the Authors ïňĄxed the sampling period from 19:00 UTC to 19:00 UTC of the following day. This choice should be explained.

Authors Response The technical detail has been reduced considerably. The sampling time started at 20:00 - now corrected. We started the instruments at 20:00 because some of the instruments were not on timers and we had to manually switch on the pumps. This was done at that time each day as it was the most practical for the ground station team. This text has now been added to explain why we started sampling at 20:00 UTC. "The DGS sampling was scheduled for 20:00 UTC (16:00 Atlantic Standard Time) as this was the most practical time of day for the DGS research staff to synchronize multiple instrument 24-hr sampling."

Page 4496 lines 3-6: The Authors say: "The ïňĆow is then split, with 15.0 lmin-1passing through the PM2.5 collection ïňĄlter and 1.67 lmin-1passing through the PM2.5-10 collection ïňĄlter providing a dichotomous sample of ïňĄne and coarse PM (Dabek-Zlotorzynska et al., 2011)" but they only discuss the PM2.5 fraction. Why? In my opinion, it should be interesting to evaluate what happens both in coarse and ïňĄne fractions and compare the results obtained.

Authors Response While the PM10/coarse fraction would be interesting, the focus of this paper is the source apportionment of PM2.5. In addition, there were not sufficient chemical species on the coarse filter to conduct accurate source apportionment that would be comparable with PM2.5 chemical species data set. The PM10 and PM coarse mass concentration descriptive statistics are contained in the BORTAS overview paper however (Palmer et al 2013, ACP, BORTAS Special Issue).

Page 4497 line 4 "14 of the measured elements , were not detected in any of the samples": this result, at least for some elements such as Pb, Cd, is quite unusual.

Could the Authors try to explain this ïňĄnding here or in another paragraph? 3.

Authors Response PM2.5 concentrations in Halifax are very low, e.g. typical means of 4 ug/m3. Not much industry or traffic. Un-leaded fuel, hence their absence.

Models Page 4500, line 4: The Authors should explain why they used 2-day back trajectories. Would longer back-trajectories have inïňĆuenced the results? Page 4500, lines 8-22:

Authors Response We ran 10-day and 5-day trajectories. The 2-day told the same story in-terms of identifying upwind source regions and provides a better visualization of the source regions in the Figure. We also ran FLEXPART to confirm the HYSPLIT trajectories (out of the scope of this paper). We have now added text to say that we ran 10-day and 5-day trajectories but for the purposes of visualization we showed the 2-day trajectories as these adequately identified the upwind source regions.

Please move the sentences indicated in the "results and discussion" section. Page 4500, line 23 and following: Too many details are provided for the United States Environmental Protection Agency (US EPA) Positive Matrix Factorization (PMF) receptor model. This method is well-known and widely applied in this type of studies, so it is not necessary to explain it with so many details. The Authors should shorten this part leaving only the details concerning the choices they made when they applied this method. 4.

Authors Response This section has been significantly reduced as suggested.

Results and discussion General comment: the discussion is very confused and hard to follow. It is very difiňĄcult to understand what contributions the Authors considered local or long-range transport-related. This part needs a deep revision.

Authors Response The choice of local and long-range sources is based upon prior knowledge and referenced work done in the region and else where. We have added a figure showing the PMF chemical species factor profiles that were used to identify the

PM2.5 source. We feel that this, together with the other reviewers comments make this section strong.

We have also added a new figure (attached) to better articulate the choice of sources observed within the seven factor profiles.

Table 1: An outlier value of temperature was reported in table 1. Why didn't the Authors leave it out of the statistical analyses? How did they evaluate the outliers? Did they ïňĄnd only this outlier?

Authors Response The obvious temperature outlier has been removed and replaced by the maximum temperature within three standard deviations of the mean. The determination of other outliers was by robust regression analysis ($\sim$ 3 standard deviations from the mean). No further outliers were found.

Page 4505 line 21: Why did the Authors consider Se and Pb as indicative of LTR pollution? Authors Response It is well known that Se is a marker for coal burning and Pb is a marker of industry and is elevated in the NE US. These two elements are often present in PM2.5 that is associated with LRT from that region to Nova Scotia. We have added the Pb marker to the sentence earlier in the document "Selenium is often used as a good marker for coal combustion with Pb acting as a good marker for industrial emissions (Chow et al., 2004; Dabek et al., 2011)." We have provided the Chow reference and Dabek et al 2011 reference again to support this statement at Page 4505 line 21 as suggested.

Please, provide some references Page 4510 lines 9-20: I suggest moving the sentences indicated in Models paragraph 5.Conclusion Page 4510:

Authors Response We have moved this section to the methods as requested (also requested by the other reviewers). We have provided the following reference for the RMSE as requested. Laupsa, H., Denby, B., Larssen, S., Schaug, J., 2009. Source apportionment of particulate matter (PM2.5) in an urban area using dispersion, recep-

tor and inverse modelling. Atmospheric Environment 43, 4733-4744.

Palmer et al.(Atmos.,Chem.,Phys.,Discuss.,12,4127-4181,2013) report “The timing of the BORTAS-B experiment during 12 July–3 August 2011 reïňĆects the climatological maximum of burning over this geographical region” and “ Halifax, Nova Scotia was one of the climatological loci of burning outïňĆow over eastern Canada”. How can the authors explain that during their measure campaign no impact is found at the surface in Halifax regarding the Boreal wildïňĄre smoke plumes? In particular, can their result be justiïňĄed only because of the low PM2.5 amount measured ? Please, explain it thoroughly.

Authors Response For the PMF modelling in this paper we did not have a unique biomass marker available. The concentrations of wild fire smoke reaching the surface are so negligible after dispersing 1000km across North America. We and the other BORTAS researchers were not just interested in the wild fire PM2.5 but also bulk chemistry of the aerosol in an effort to understand chemical transport reactions occurring in the air parcels as the tracked across the region. In addition, the other BORTAS researchers taking measurements at the Dalhousie Ground station were also interested in the PM2.5 chemical speciation and sources to help explain their results.

Technical corrections Page. 4495, lines 15-16: Please, provide the list of chemical species measured and reported in brackets following the alphabetical order.

Authors Response The list is provided and placed in alphabetical order

Page 4495 line 8: the information on the sampling period is confused. Please, specify better when the sampling stopped on 11/12 July. On this day, how many hours did the ïňĄlter sample?

Authors Response The text has been changed as below to clear up any confusion. A 24-hour filter sample was collected at the BORTAS-B DGS from 20:00 UTC on 11 July 2011 to 20:00 UTC the following day. Uninterrupted 24-hr filter samples were then

taken from 20:00 UTC on 13 July 2011 to 20:00 UTC on 26 August 2011 resulting in 44 consecutive days of PM2.5 samples. A total of 45 filter sampling days. Continuous measurements of PM2.5 mass concentration, black carbon (BC), organic matter (OM) and meteorology were also collected over the same sampling period. The other collocated measurements at the DGS that are not featured in this paper are described in Palmer et al. (2013).

Page 4498 line 14: the sentence contains a misprint: the term “organic” was repeated twice.

Authors Response Corrected.

Page 4498 line 27: the Authors say: “Meteorological data at the BORTAS-B DGS was collected every 15min using a Davis Vantage Pro II weather station” but the caption of Table 1 reports “Descriptive statistics for the meteorological variables obtained at the DGS during the PM2.5 sampling period based upon 5-min average data”. Please, check.

Authors Response Table 1 should be 15-min. Now corrected.

Page 4499 lines 6-8: Note that ïňĄgures must be numbered consecutively as they appear in the text. As a consequence, it is quite unusual to ïňĄnd in the text ïňĄgure 9 after ïňĄgure 1 and before ïňĄgure 2.

Author response The figure captions in the text have been placed in a more logical order.

Please, move this sentence to the “results and discussion” paragraph. Page 4500, line 5:

Authors Response This sentence/paragraph has been modified based upon the other referees similar request.

Please, check the sentence. Page 4503 lines 1-2: The Authors say “Potassium is our

preferred marker of long-range wildïňĄre smoke plumes as it is conserved from source to receptor". Please provide a reference for this statement.

Authors Response This section has been modified and a reference added as requested (Ward et al., 2012).

Page 4504 lines 5-8: Please, revise the sentence that appears badframed in its ïňĄrst part.

Authors Response Change as follows: "The HYSPLIT 2-day air mass back trajectories provided in Figure 2 shows that 40% of the air masses entering Halifax. . .."

Page 4504, line 22. The NAPS abbreviation must be deïňĄned.

Authors Response Now defined earlier in the manuscript upon first instance

Figure 2 caption: The Authors said: "Two trajectories were obtained for each 24-h sampling period (07:00UTC and 19:00 UTC)" (page 4500, lines 5.7) but the caption of ïňĄgure 2 reports that trajectories were initialized 08:00UTC. Please, check and rectify.

Authors Response Now rectified as follows: Figure 2. Map of ensemble HYSPLIT 2-day air mass back trajectories between 11 July 2011 and 25 August 2011. Trajectories were initialized twice per day at 08:00 UTC and 20:00 UTC with an arrival height of 500 m. Colours denote upwind source region (cyan = Marine, red = SW, green = WNW and blue = N)

Figure 3 reports the HYSPLIT 2-day air mass back trajectory vertical proïňĄles but it does not specify the back-trajectories they refer to (i.e., 07:00UTC or 19:00 UTC).

Authors Response Change made as follows: Figure 3. HYSPLIT 2-day air mass back trajectory vertical profiles initialized twice per day at 08:00 UTC and 20:00 UTC

Page 4519, Table 1: the number of observations of the meteorological parameters is 42, while those related to PM2.5 are 45. Why? Please provide a brief explanation in the text.

Authors Response It was a typo. It should be 45. Correction made

New Figure Captions

Figure 1. Location of the DGS used during BORTAS-B (source of maps: free within ArcGIS v10)

Figure 2. Map of ensemble HYSPLIT 2-day air mass back trajectories between 11 July 2011 and 25 August 2011. Trajectories were initialized twice per day at 08:00 UTC and 20:00 UTC with an arrival height of 500 m. Colours denote upwind source region (cyan = Marine, red = SW, green = WNW and blue = N)

Figure 3. HYSPLIT 2-day air mass back trajectory vertical profiles initialized twice per day at 08:00 UTC and 20:00 UTC

Figure 4. Time series of total PM2.5 mass and major species concentration

Figure 5. Time series of total PM2.5 mass and macro species concentration

Figure 6. Time series of total PM2.5 mass and micro species concentration

Figure 7. Time series of total PM2.5 mass and trace species concentration

Figure 8. Source profiles for the seven PMF factors

Figure 9. Time series of PM2.5 source apportionment based upon PMF output

Figure 10. Source Contribution Rose

Figure 11. Back trajectories associated with the highest values of each PMF cluster.

Figure 12. Average mass concentration (μg/m3) of attributed sources and percentage source contributions over the 45 days of sampling

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C2654/2013/acpd-13-C2654-2013-supplement.pdf

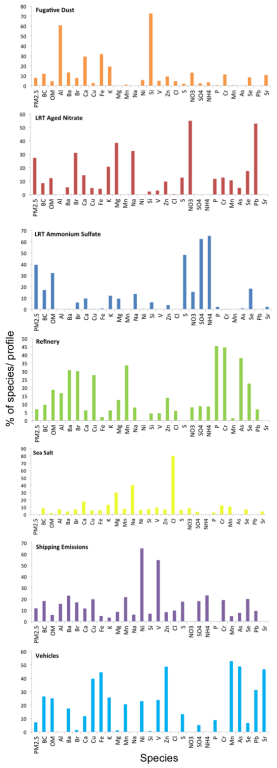

Fig. 1. Figure 8. Source profiles for the seven PMF factors