We thank the two anonymous referees for their insightful comments to the manuscript and helpful suggestions for improving the presentation quality. Below, we explain how the comments and suggestions are addressed (our point-by-point responses in blue) and make note of the changes that have been made to the manuscript, attempting to take into account all the comments raised by both referees.

Referee #1

General comments:

An analysis of model results for BC in snow in the Northwestern USA from simulations with CAM5 is presented. The focus of the paper is the validation of model results based on a combination of a large number of high-quality observational data sets. As a novelty, a Positive Matrix Factorization (PMF) analysis is performed to determine biomass and fossil fuel sources of BC in the snow.
Many models produce substantial biases in simulated BC concentrations in the atmosphere in this region. An analysis of the relationship between BC in the atmosphere and deposition on snow is a very useful approach with regard to needed improvements of climate and air quality models. Unfortunately, there are several key aspects of the approach that seem problematic. In particular, the approach likely underestimates the influence of biofuel emissions in the model as explained in more detail in the following. Second, comparisons between BC concentrations in snow and air are based on unverified assumptions about correlations between these quantities.

Response: please see our responses to the more specific comments.

Specific Comments:

Page 12964, line 7 - 13: Please clarify whether sensible and latent heat fluxes are specified in calculations of atmospheric properties and land surface processes. How do amounts of snow and BC processes in snow in specified dynamics mode compare with results from the freely running model and how accurate are results? It seems that this approach has previously been used to study atmospheric processes but it is not obvious how well it works for snow and BCC.
Response: In the specified dynamics mode, surface sensible and latent heat fluxes are specified in calculations of atmospheric properties but not the land surface processes. However, precipitation (including rain and snow) and BC deposition to snow are calculated in the atmospheric component of the model. With the constrained meteorological fields the model can simulate clouds, precipitation and aerosol processes better than in the freely running mode (e.g., Ma et al., 2013), especially, for

a specific time period (as opposed to climatology). As shown in our Figure S5 (in the supplement), the CAM5 simulated snow cover fraction (SCF) has a good agreement with satellite retrievals. The three-month mean SCF for CAM5 is 50% over Northwest USA and 99% over West Canada, comparing to mean SCF from MODIS of 58% over Northwest USA and 96% over West Canada. This is the exact reason why we choose to run the model in specified dynamics mode for comparing with the field measurements made during January-March of 2013. We have now clarified more on this in the revised manuscript.

Page 12964, line 26: The yet unpublished ECLIPSE data set is not properly acknowledged. See the ECLIPSE website for details.
Response: Thank you for pointing this out. We now cite Stohl et al. (2015) in the text, as suggested by referee #2, and have added the following statement to the acknowledgment: "ECLIPSE emission data sets are available from http://www.geiacenter.org/access. Funding for the development of the ECLIPSE emission data set was provided by the European Union Seventh Framework Program (FP7/2007–2013) under grant agreement no. 282688 – ECLIPSE.".

Page 12965, line 5-8: It seems highly problematic to apply the ratio of biofuel to total emissions from the old AEROCOM/GFED emission data set by Dentener et al. (2006) to the new combined ECLIPSE/GFED3 data set that is used in CAM5. This will likely lead to incorrect estimates of fossil fuel and biofuel emissions. Different emission sectors are considered in these data sets (e.g. oil and gas flaring emissions are included in the ECLIPSE data set but are not included in the AEROCOM data set). There are also substantial differences in emissions from sources that are common to both data sets. For GFED3, there is a 43% increase in emissions for boreal North America compared to GFED2 (van der Werf et al., 2010). The latter implies that biofuel emissions and contributions to BC in snow in North America are substantially underestimated with this approach, which likely explains diagnosed underestimates in BB contributions to BC in snow in CAM5 in Fig. 6, a key conclusion.
Response: First of all, we would like to clarify on a possible misunderstanding here. When apportioning the ELCIPSE emissions to fossil fuel and biofuel, we did not use fire emission data sets (i.e., GFED2 or GFED3). The difference between GFED3 and GFED2 data sets, which are both attributed to biomass burning emissions, would not directly affect the calculation of biofuel emissions and contributions since the apportionments of ECLIPSE emissions did not use GFED3 data sets. On the other hand, the 43% increase in GFED3 emissions for boreal North America that the referee pointed out does not appear in the JFM mean emissions we used in our simulation. In

the following Table R1, we compare JFM mean emissions from the different source regions/sectors between the ECLIPSE/GFED3 and IPCC-AR5/GFED2 data sets. Biomass burning (fire) emissions in North America are not so different between the two data sets, and much smaller than fossil fuel and biofuel emissions.

We agree that the additional oil and gas flaring emissions in the ECLIPSE data set would affect the apportionments. They are substantial in the Arctic (e.g., ARC) and less so in Canada (e.g., WCA and ECA), but somehow the ECLIPSE data set has even lower emissions than the AR5 data set over USA (e.g., NEU, SEU, NWU and SWU), which might partly explain the overall low bias in the modeled BCC concentrations. However, we don't have observations to evaluate against. It is also worth noting that the difference in global total JFM emissions between the two data sets (7.692 vs. 7.718 Tg yr^{-1}) is very minimal. To give a better idea on how the apportionments of FF vs. BF might affect the source attribution and facilitate a comparison with other emissions data sets, we have revised the Figure 1 to separate out BF from the BB category. The figure is shown below (Figure R1).

Table R1: January-February-March (JFM) mean emission (Tg yr^{-1}) from each source region for two different inventories (i.e., ECLIPSE/GFED3 vs. AR5/GFED2).

Source region	Fossil fuel (Tg yr^{-1})			Biofuel (Tg yr^{-1})			Biomass burning (Tg yr^{-1})		
	ECLIPSE	AR5	Diff	ECLIPSE	AR5	Diff	GFED3	GFED2	Diff
ARC	0.024	0.003	0.021	0.003	0.000	0.003	0.000	0.000	0.000
WCA	0.015	0.014	0.001	0.002	0.002	0.000	0.000	0.002	-0.002
ECA	0.012	0.008	0.004	0.003	0.002	0.001	0.000	0.000	0.000
LAM	0.273	0.321	-0.048	0.106	0.113	-0.007	0.051	0.206	-0.155
NEU	0.096	0.167	-0.071	0.025	0.042	-0.017	0.001	0.002	-0.001
SEU	0.055	0.082	-0.027	0.011	0.017	-0.006	0.006	0.004	0.002
NWU	0.012	0.019	-0.007	0.002	0.003	-0.001	0.000	0.000	0.000
SWU	0.027	0.047	-0.020	0.008	0.015	-0.007	0.000	0.000	0.000
EAS	1.400	1.195	0.205	0.559	0.448	0.111	0.099	0.057	0.042
SAS	0.303	0.204	0.099	0.645	0.435	0.210	0.245	0.135	0.110
SEA	0.195	0.187	0.008	0.236	0.194	0.042	0.504	0.561	-0.057
ERCA	0.582	0.734	-0.152	0.092	0.106	-0.014	0.001	0.022	-0.021
AFME	0.381	0.250	0.131	0.802	0.376	0.426	0.693	1.481	-0.788
PAN	0.030	0.037	-0.007	0.004	0.005	-0.001	0.016	0.079	-0.063
ROW	0.172	0.142	0.030	0.000	0.000	0.000	0.001	0.001	0.000
Global	3.577	3.410	0.167	2.498	1.758	0.740	1.617	2.550	-0.833

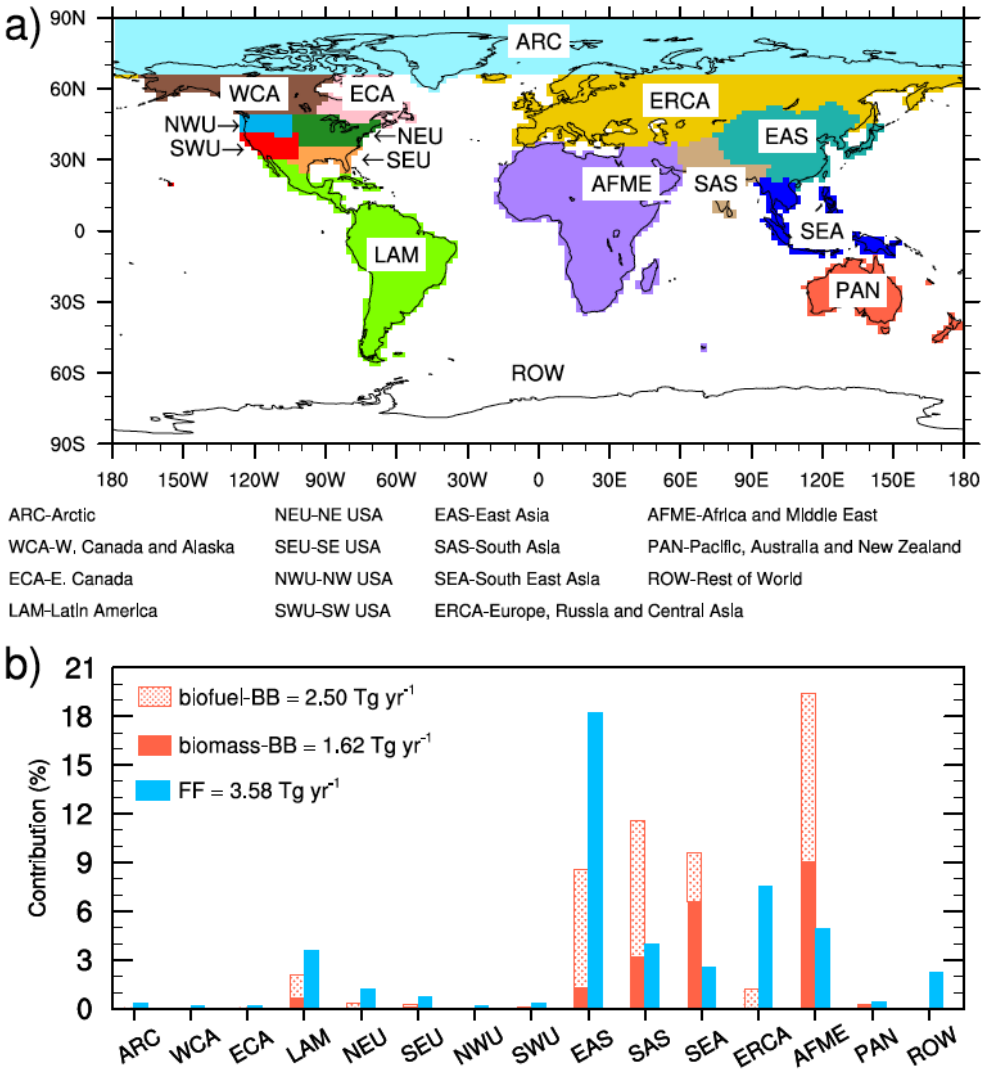

Figure R1. (a) Tagged source regions and (b) the contributions (%) to the global mean BC emissions (7.69 Tg yr^{-1}) for January, February and March from the individual source regions (marked on the horizontal axis) and sectors (FF in blue, biomass-BB in solid red, and biofuel-BB in dotted red).

Page 12968-12969: The presentation of equations and associated description of the analysis method seems somewhat lengthy and complicated. Maybe some of this could be moved to the supplement or otherwise be simplified?

Response: We agree with the referee that the equations are lengthy but they are important to calculations used in the results section. Thus we decide to move these equations in section 2.4 to the Appendix so that main text of the paper will flow better, yet readers can easily access the equations.

Page 12971-12972, section 3.2: I found it difficult to understand this section. A table of concentrations and biases in different regions would be beneficial for a more

concise summary of results.

Response: The four panels in Figure 3 visualize the model-observation comparison in different ways. We feel that the BCC concentrations and model biases have been adequately presented. Also, we mean to make the comparison for the entire regions rather than focusing on individual sites. However, following the referee's suggestion, we have made a table (Table R2), summarizing all the concentrations in Northwest USA and West Canada, and added it to the supplement as Table S3.

Table R2: BC-in-snow-column concentrations from observations and the CAM5 simulation in Northwest USA and West Canada at 36 sites

Northwest USA			West Canada		
Comparison pair i	$\overline{C_{obs}^{i}}$ (ng g^{-1})	$\overline{C_{mod}^{i}}$ (ng g^{-1})	Comparison pair i	$\overline{C_{obs}^{i}}$ (ng g^{-1})	$\overline{C_{mod}^{i}}$ (ng g^{-1})
1	8	1	21	39	44
2	15	7	22	36	18
3	25	8	23	18	19
4	31	9	24	18	13
5	29	3	25	15	7
6	52	15	26	19	6
7	78	10	27	7	4
8	88	15	28	11	13
9	62	14	29	12	16
10	45	14	30	21	22
11	35	13	31	22	29
12	28	20	32	16	25
13	34	37	33	15	9
14	17	24	34	13	20
15	19	26	35	21	27
16	18	47	36	22	30
17	73	30	mean±SD	19±8	19±11
18	110	23			
19	37	28			
20	67	40			
mean±SD	44±28	19±13			

Page 12973, lines 6-11: Comparisons between the snow column BC mixing ratio (BCC) and near-surface atmospheric concentrations of BC (BCS) are problematic for several reasons. First, at any given location, vertically integrated concentrations of BC

in the snow are largely determined by the mean deposition fluxes of BC and snow during the time period since the snow pack started to form in the fall of the previous year. If data from permanent snow fields is considered then emissions of BC from previous years may also be important. Consequently, comparisons with mean BC concentrations in air in January-February-March (JFM) should be replaced by comparisons that are based on overlapping time periods for BCC and BCS. Second, as is also pointed out in the paper, spatial variability in BC concentrations is large and cannot be fully quantified based on the relatively small number of measurement sites. The sparse distribution and lack of co-location of measurements limits the statistical robustness of the comparisons, which is not quantified. Furthermore, estimates of LMNB and LMNE are biased low in the Northwest USA region for both BCC and BCS (see previous manuscript pages). This points at a common explanation for biases in these quantities (such as an underestimate in BC emissions), opposite to the explanation given here. It is not obvious how biases in the Northwest USA region can be explained by results for Canada since the impact of local emissions on regional concentrations is so high as the study shows?

Response: None of the snow samples of Doherty et al. (2014) were of permanent snowpacks; all were from seasonal snowpacks. We agree that seasonal snowpack at our sampling sites may have started to accumulate in the fall of the previous year. However, as shown in Figure 7b of Doherty et al. (2014) there is not a vertical gradient in the mixing ratio of BC in snow at our sites. Instead there is variability, and we thought averaging across this variability would provide a more representative value for typical mixing ratios of BC in snow. The seasonality of BC emissions (i.e., biomass burning) in the cold season is also small. To address the referee's concern, we have BCC and surface-BC concentrations in four individual winter months listed in Table R3 (added to the supplement as Table S1). There is no clear trend of surface-BC and BCC variation from December to March, while for the same month surface-BC and BCC concentrations are strongly correlated. We have explained in Section 2.1 why BCC is used instead of surface-BC in the model evaluation.

Note that we are not directly comparing atmospheric concentrations (BCS) versus in snow (BCC), but rather the differences in the model biases in BCC and BCS. Regarding the sparseness of sites: We have a distribution of sampling locations across the regions for both BCC and BCS, so while the comparison is not perfect it is nonetheless of value. To address the referee's concern we have added text noting that the measures of BCC and BCS are from different locations and are not necessarily representative of the whole model grid box, so the comparison is not ideal but is nonetheless informative.

Finally, note that we do not exclude an underestimate of BC emissions as a cause of the difference; we simply also note that there is another possible cause of the low bias in BCC. We don't see a conflict here. Indeed, we make the point that any potential bias caused by model representation of BC deposition processes should show up in the comparison of BCC in both regions consistently, rather than just in the Northwest USA. Thus, we hypothesize that the difference in model bias in BCC vs. BCS is likely due mostly to an error in model emissions. The text has been edited to more clearly state this.

Table R3: CAM5 Monthly BC-in-snow-surface and BC-in-snow-column at 36 sites from December to March. ("N/A" means no snow in the model grid.)

Comparison pair *i*	BC-in-snow-surface (ng g^{-1})				BC-in-snow-column (ng g^{-1})			
	Dec	Jan	Feb	Mar	Dec	Jan	Feb	Mar
1	13	4	N/A	N/A	10	1	0	0
2	7	22	7	1	5	9	5	0
3	9	20	6	N/A	10	13	4	0
4	6	20	7	N/A	4	13	6	0
5	6	9	2	N/A	5	6	1	0
6	6	12	24	N/A	5	9	20	0
7	7	22	6	N/A	6	16	4	0
8	14	13	20	57	13	11	13	21
9	11	15	20	54	12	11	13	19
10	11	19	30	49	8	9	13	20
11	9	16	22	35	8	9	12	17
12	24	22	37	35	20	17	18	26
13	84	56	48	42	57	59	25	27
14	39	57	22	17	26	46	12	14
15	51	87	42	27	21	36	25	18
16	86	56	44	61	53	61	41	38
17	43	27	40	59	27	26	27	39
18	32	28	35	37	25	23	23	24
19	38	39	37	42	30	29	28	26
20	76	51	41	59	59	47	39	35
21	74	48	70	73	50	47	43	43
22	30	19	49	37	16	17	18	20
23	32	26	46	46	14	17	18	21
24	16	14	32	38	12	11	12	14
25	6	6	20	18	7	6	7	8
26	5	5	12	10	5	5	6	6
27	2	2	5	4	5	4	4	4
28	16	14	32	38	12	11	12	14
29	20	14	35	49	16	15	15	18

30	30	29	44	54	16	20	22	24
31	57	37	37	41	48	32	28	28
32	34	38	29	41	24	26	24	25
33	9	8	22	32	8	8	9	10
34	25	29	32	47	18	20	19	21
35	31	40	34	46	27	29	26	27
36	37	44	44	44	27	28	31	30

Page 12974, lines 7-28: Potential emissions of BC particles from soils as source of missing BC in snow in the model is an interesting topic. Soils can indeed contain microscopic particles of charcoal from vegetation fires and particles from coal combustion (Schmidt and Noack, 2000). However, various processes such as soil erosion, BC decomposition, etc. need to be considered for potential emissions of BC from soils. What concrete observational evidence exists for a soil source of measured BC snow concentrations in this study? How can the fingerprint of a soil component in the PMF analysis be explained? Soil particles and BC are both often found in snow but this does not necessarily imply a common source. For instance, deposition of soil and BC to a snow field would be positively correlated if disturbed soils and fossil fuel sources of BC are both upwind of the snow field. Further, forest fires plumes may contain soil chemical elements and can therefore also produce a positive correlation. Hence it is not clear how a lack of BC in snow can be explained by missing (direct) emissions from soils in the model.

Response: We too think this is an interesting topic and thus we explained in the text our hypothesis of the importance of soil BC in the Northwest USA domain (e.g., Schmidt and Noack, 2000; Hegarty et al., 2011). We do not have concrete and direct observational evidence of there being BC in the soil source that ends up in the snow, but infer this from the PMF analysis. The interpretation of the soil factor in the PMF analysis is based on the measured high loadings of well-known soil constituents such as Al, K, Ti, V, Ca and As (see Figure S3 in the supplement). We agree that some of these soil chemical elements may partly originate from forest fires. A PMF analysis is much more than a simple correlation analysis. The ranked, orthogonal covariance analysis tells us that the optimum variance reduction is achieved when part of the BC is in fact from a source distinct from direct fire emissions even when such emissions are present. Furthermore, this source is associated with soil markers. Hence, the PMF results do suggest a separate soil source and such a source is both plausible and consistent with the soil data for the Northwest USA region, as discussed in the text.

While the magnitude of this source of BC to snow as quantified by the PMF analysis has large uncertainties, it does suggest that this mechanism for getting BC into snow is not insignificant in some locations. We believe that wind-blown soil BC, as

opposed to atmospheric BC directly from emissions, contributes to BC measured in the snow. Importantly, as we point out, this process is not considered in the model simulation. We have now clarified more on this in the revised manuscript accordingly.

P. 12978, line 5-7: Please add more quantitative information about the differences. What are the mean values and standard deviations?

Response: Done as suggested. The text is revised to "Compared to the original PMF values (including contributions from FF, BB and soil), CAM5 underestimates the BB contribution for 80% of the comparison pairs (modeled mean and standard deviation of 18%±5% vs. PMF values of 28%±22%) and overestimates the FF contribution for all comparison pairs (82%±5% vs. 47%±21%)."

P. 12978, line 9: Define what combustion sources are considered. Does this refer to fossil fuel combustion emissions (P. 12974, line 9)?

Response: The combustion sources mentioned here include fossil fuel combustion and biofuel/biomass burning that are considered in the CAM5 simulation. This has now been clarified in the revised manuscript.

P. 12980, line 4-5. A simple linear relationship in latitudinal variations in BC radiative forcing and BC deposition flux cannot necessarily be expected and the meaning of such a relationship is not clear. For instance, the radiative forcing depends on insolation and therefore latitude, which is not considered here. In addition, as explained above, JFM deposition fluxes and concentrations are not a good proxy of the BC loading in the snow pack. Furthermore, the discussion of radiative forcings does not seem to be logically connected to discussions in the rest of the paper.

Response: We agree with the referee's arguments here. However, both the atmospheric and in-snow BC radiative forcings were calculated interactively in the CAM5 simulation using online radiative transfer models with factors such as atmospheric and in-snow BC concentrations, latitude-dependent insolation, particle sizes and optical properties considered. No simple linear relationship between BC forcing and deposition flux was used. Such a relationship was simply meant as a first-order approximation if we were to attribute the calculated radiative forcing to the different sources. This has now been clarified in the revised manuscript. We believe the radiative forcing calculation would be of interest to colleagues who have done/are doing similar calculations for the same region and other parts of the globe. It is also useful to compare the forcing between different regions and with global mean values. Therefore, we decided to keep the discussion but we now provide some context for this in the introduction section.

Referee #2

This paper concerns a tagging technique of black carbon (BC) emissions to study the source-receptor relationship for BC in the atmosphere and on snow in Western North America using CAM5. The model results are compared with observations in the region. As most models seems to underestimate BC near the surface at high latitudes, this paper is relevant and might be of great interest to the scientific community. The questions raised in the study is within the scope of ACP. The figures are in good quality and the figure captions explain the figures well. However, for the paper to be published in ACP, some revisions need to be done. The authors should work more on the overall presentation of their results.

Specific Comments:

1. In general, I think the paper is somewhat too long and unfocused. The paper would benefit from a substantial reduction both in the Methods chapter and also the Results and Discussion. An effort to focus those parts would make the paper much easier to read. You explain your methods well, but there are still parts in Methods that can be moved to Introduction or Supplementary. For instance, the second paragraph in 2.1 Observations you discuss what is new in this study and what previous studies have done. 2.1 should only describe the actual observations, and the rest can be skipped/compressed/moved to introduction. There are many equations in 2.3 and 2.4 that can be moved to Supplementary if necessary. Also, in the Results chapter, only results should be presented (and not repetition of Methods for instance). The whole Results chapter can be shortened for clarity.

Response: Thanks for the constructive suggestions. We have now significantly revised the Methods section and the Results section following the suggestions. Section 2.1 is shortened with some necessary information moved to introduction. Some of the equations in sections 2.3 and 2.4 are lengthy but they are important to calculations used in the Results section. Thus we decide to move them to the Appendix so that they won't affect the flow of the paper but can be easily accessed by readers. Repetition of methods in the Results section has been removed.

2. The authors should explicitly state why this study is important. Also, what are the benefits of using this tagging technique instead of doing emissions perturbations? By focusing the paper and skipping parts that are not relevant, the new contribution would be easier to detect.

Response: Black carbon (BC) is believed to be an important climate-warming forcing agent in the climate system. However, the global BC forcing estimate is very uncertain, mostly because of large uncertainties in global BC emissions and

parameterizations of BC-related processes in global models. Observational and modeling studies focusing on specific regions have been useful for reducing such uncertainties. Previous BC studies, especially those addressing BC-in-snow effects, have mostly focused on polar regions and mountainous regions. The climate effect of BC might be greater in mid-latitude regions, among which North America has received less attention. The recent large-area survey of observed BC in snow in Western North America provides an opportunity for assessing how well global models predict BC concentrations in snow. These factors motivated the present study. Another important reason is that the prediction of global spatial distribution of BC by the Community Atmosphere Model (CAM5) has been significantly improved (Wang et al., 2013) and, additionally, a BC source-tagging technique was recently implemented to the model for source-attribution study (Wang et al., 2014; Zhang et al., 2015). Different global modeling approaches have been previously employed to establish aerosol source-receptor relationships, among which emissions perturbations have been widely used. Not only does this approach assume a linear response to perturbations to get fractional contribution of different sources, but it also requires additional simulations for each source perturbation. The latter would add about 30 times more computational cost that our direct tagging approach for the 32 source regions/sectors. Thus we believe the tagging technique is more computationally efficient and gives more accurate results.

We have now clarified this in the revised manuscript.

3. I am a bit confused why you separate out BF from FF and lump together with BB. The ECLIPSE emissions have different sectors compared to the previous ones from Dentener et al (2006). For instance flaring is included as a sector in ECLIPSE. There are no easy way to separate out BF, but you should at least discuss your assumptions further. What uncertainties do you introduce?

Response: The main purpose of the regrouping is to facilitate the comparison with the PMF analysis, which is able to distinguish BC from the combustion of fossil fuels versus BC from the combustion of biomass/biofuels. The chemical markers from open biomass burning (e.g. forest fires) and biofuel burning (e.g. woodsmoke from fireplaces and wood stoves) are quite similar so we could not distinguish the two in the PMF. The text has been edited to more clearly state this. We also revised Figure 1 to show BF and biomass burning contributions separately in each source region, and created a table (Table R1) to compare emissions from the different sectors between ECLIPSE/GFED3 and the popularly used IPCC AR5 data sets, which indicates that the oil and gas flaring in ECLIPSE should not significantly affect our results. Please see our response to the similar comment from referee #1.

4. Klimont et al. is still in preparation the ECLIPSE emissions data set v4a, but in the meantime Stohl et al. 2015 should be a sufficient reference: http://www.atmoschem-phys-discuss.net/15/15155/2015/acpd-15-15155-2015.html Here, the emissions are described in more detail.

Response: Thanks for pointing to the reference, which is now referred to in the manuscript.

5. In the results chapter; would it be an idea to not use the abbreviations for the source regions? This will make it easier to follow.

Response: We now tried to spell out the source regions frequently and make the results easier to follow.

6. The radiative forcing sub chapter was unexpected. You have not mentioned this earlier in the paper. How is the forcing calculated? How did you estimate BC DRF in the atmosphere? As a difference between surface and TOA? How do you calculate the surface RF (dimming) compared to output from SNICAR? Also, you conclude that a positive forcing at the surface (?) means heating at the surface. This is not correct, and I would avoid writing this unless you have a fully coupled climate run. Whether BC warms the surface depend on the height of BC in the atmosphere. You also find a correlation between deposition and surface RF. What about the albedo? Solar radiation? You only look at the winter months. The section in its current form seems misplaced. I suggest to either expand the analysis, or to skip this section entirely.

Response: We have now added to the introduction some background information for the BC radiative forcing calculation. Although our model simulation is not a fully coupled climate run, in which temperatures of ocean, land surface and atmosphere evolve freely, both the atmospheric and in-snow BC radiative forcings were calculated in the CAM5 simulation using online radiative transfer models (RRTMG for atmospheric radiation and SNICAR for BC in snow/ice). BC direct radiative forcing (DRF) in the atmosphere was estimated as the difference between the net radiative fluxes at the surface and TOA. Atmospheric BC has a net heating effect in the atmosphere and a net cooling effect at the surface (i.e., surface dimming). BC-in-snow effect is to increase the absorption of solar radiation in snow and, therefore, reduce the reflected radiation from the surface, representing a radiative heating effect. In the surface energy budget equation, atmospheric BC reduces downwelling shortwave radiative flux while in-snow BC reduces upwelling shortwave radiative flux. We believe there is no problem in this. However, the longwave radiative fluxes and surface sensitive/latent heat fluxes are not discussed here.

7. I'm curious about the BC in soils. How do you find this in your own analysis? I am not sure if I understood this correctly. Since this is part of your conclusions, it should be elaborated a bit more I think.

Response: BC in soil was identified by chemical "fingerprints' in the PMF analysis (Figure S3; more detailed discussion of the PMF analysis can be found in Doherty et al., 2014). Please also see our response to the similar comment raised by referee #1.

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