Anonymous Referee# 1

This manuscript reported the data of light absorbing particles (LAP, such BC, OC and dust) measured in snow/ice in northern Pakistan and estimated the induced snow albedo reduction and corresponded radiative forcing during 2015-2016. Authors found the concentration of BC, OC and dust in aged snow is higher than in fresh snow and ice and the concentration over northern Pakistan is higher than over the Himalayas and Tibetan Plateau. Estimated LAP-induced daily mean snow albedo reduction is approximately 0.07-12.0% and corresponded radiative forcing is approximately 0.16-43.5 Wm-2, depending on snow type, solar zenith angle, and locations. Also different methods are used in this study to identify the source regions of pollutants measured in this region.

Overall the results are interesting and measurement data are valuable for the community. The figures and tables in the manuscript are relevant, but not very good in quality, and need to be improved. In general the paper is well written but in many places the English could and should be improved. There are some major weaknesses in the manuscript, especially in the source region identification part (3.5) and aerosol type frequency distribution part (3.2). After the below comments are appropriately addressed, I would suggest to accept the manuscript for publication in the Atmospheric Chemistry and Physics.

Response:
We thank the reviewer for their comments that significantly contributed to improving the original manuscript. Please see below our comment-by comment-responses to each of the reviewer's comments and suggestions.

Reviewer Comments in black

Responses in blue

Modified text in the revised manuscript is in green.

Technical comments:

Introduction, I would suggest use Light-absorbing particles (LAP) instead of Light absorbing impurities, see Qian et al., 2015, which by the way is a review article for both measurement methods and modeling activities. This article is so relevant so probably should be cited in Introduction part.

Response:

Light-absorbing particles (LAP) has been used instead of Light absorbing impurities (lines 14,48,82,89,151,292,418,420, and 453). We changed the title accordingly. The recommended article has been cited in the introduction (lines 83). The text has been quoted below for your reference.

"A number of authors have described the concentration and impacts of light absorbing particles in the Tibetan glaciers (for example Qian et al., 2015; Wang et al., 2015; Que et al., 2016; Zhang et al., 2017; Li et al., 2017; Niu et al., 2017)."

Lines 62-65, Besides warming efficiency, another important characteristic of LAPs is its higher snowmelt efficiency, see Qian et al., 2011.

Response:
An additional sentence related to warming has been added in the revised manuscript (lines 67-69).

"Besides warming efficiency, another important characteristic of BC is its higher snowmelt efficiency. The snowmelt efficacy induced by BC in snow is larger for snow cover fraction and snow water equivalent than induced by carbon dioxide increase (Qian et al., 2011)."

Lines 110, precipitation 0.412 +/- 2 mm for per day or per year?

Response:
The sentences have been modified (lines 117-119), given below for your reference.

"According to the 10 years record (1999–2008) of the two nearby climatic stations, the mean total annual precipitation was 170 mm at Khunjerab (36.83°N, 75.40°E, 4730 m) station, and 680 mm at Naltar (36.29°N, 74.12°E,2858 m) station,"

Lines 123-128, again sample method is summarized in Qian et al., 2015.

Response:
We have referred to Qian et al., 2015 in the methodology section (line 138).

"Qian et al., 2015 summarized sample methods for light absorbing particles in snow and ice from different region including Arctic, Tibetan Plateau and mid-latitude regions."

Section 2.3.1, without 2.3.2, really needs 2.3.1? More details are needed regarding how the L-2 data are processed.

Response:
As per reviewer suggestions, the title/ heading of section 2.3.1 has been deleted (line 162). This part (paragraph) describes the aerosol subtypes, and CALIPSO level 2 lidar data processing.

We have also added details about L-2 data processing in the modified manuscript as quoted below (lines 163, 169-174).

"The CALIPSO models define aerosol subtypes, with 532-nm (1064 nm) extinction-to-backscatter ratio. The CALIPSO Level 2 lidar vertical feature mask data product describes the vertical and horizontal distribution of clouds and aerosol layers (downloaded from https://eosweb.larc.nasa.gov/project/calipso/aerosol_profile_table). On the basis of observed backscatter strength and depolarization, the aerosol subtypes have been pre classified in the downloaded data. The details of algorithm used for the classification have been presented in Omar et al., 2009. Percentage contribution of individual aerosol subtypes were plotted using Matlab."

The number of counts for a specific aerosol type in specific month were plotted as indicated in Figure 2 and Figure S4. According to our understanding, few authors in the past (including Cong et al., 2015; Ali H. Omar et al., 2009 and Wang et al., 2016) used the sub-type aerosol data.

Lines 155-159, This paragraph should be removed or moved to Introduction section.

Response:
The paragraph has been moved to introduction section with few modifications as suggested by the reviewer (lines 47-49 of modified manuscript). The whole sentence is given below for your reference.

"However, the exact amount of albedo reduction also depends on the refractive index, snow age, grain size, solar zenith angle (SZA), snow density, dust particle size and concentration, particle morphology, surface roughness, snow depth, liquid water content, snow shape and topography (Wiscombe and Warren 1985)."

Section 2.5 is very poorly organized and kind of just present whatever tools you have or used before, without a clear goal or coherence in science structure. Must be rewritten.

Response:
The section has been reorganized and edited by a native English speaking editor (lines 210-269). The modified section has been quoted below for your reference.

2.5 Source regions of pollutants
Three methods were used to identify the potential source regions of pollutants found at the study site: wind maps, emissions inventory coupled with back trajectories, and a region-tagged chemical transport modeling analysis.

Wind vector maps were prepared using MERRA-2 reanalysis data (available from the National Aeronautics and Space Administration [NASA] https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/docs/). The U and V wind components were combined into a matrix around the study area for each individual month and then plotted against latitude/longitude values to show the spatial variance of monthly wind stress at 850 mb using arrows to indicate the direction and intensity of wind.

Air trajectories were calculated backwards from the sampling sites (S1: 36.40°N 74.50°E; S6: 35.46°N 72.54°E) to identify potential source regions for the pollutants using the web version of the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT-4) model (Draxler and Hess, 1998). The HYSPLIT-4 model has been used by others to compute air mass trajectories to identify possible source regions (Ming et al., 2009; Zhang et al., 2013). Reanalysis meteorological data from the same source as the wind data (https://www.esrl.noaa.gov/psd/data) were used as input data in the HYSPLIT model for May, June, and December 2015, and January 2016. HYSPLIT was run in a seven-day backward trajectory mode with trajectories initiating every six hours (0, 6, 12, and 18) on a daily basis from 4 May to 19 June 2015 (77 days during summer) and from 1 December 2015 to 31 January 2016 (62 during winter). The HYSPLIT model results were combined with Representative Concentration Pathways (RCPs) emission data for 2010 (available from http://sedac.ipcc-data.org/ddc/ar5_scenario_process/RCPs.html) to identify the source location. The data file used as a RCP emission inventory was "RCPs_anthro_BC_2005-2100_95371.nc". This comprises emission pathways starting from identical base year (2000) for multiple pollutants including black carbon and organic carbon. According to the description of the file, biomass burning sources were included in the RCP emission inventory that were utilized with the back-trajectory analysis. RCP had the same emissions sectors as for Hemispheric Transport Air Pollution (HTAP) emission inventory used in the molding part. The emission sectors include fuel combustion, industries, agriculture and livestock. The difference in HTAP and RCP inventories is the resolution. HTAP had relatively high resolution (0.1 x 0.1 degree) as compared to RCP (0.5 x 0.5 degree). Some discussion related to the inventory and the sectorial detail (12 sectors), which was used for the base year calibration of the RCPs is given in Lamarque et al., 2010. Monthly CALIPSO satellite based extinction data from 2006 to 2014 were used to calculate the vertical profile for aerosol extinction over the study region. The CALIPSO extinction profile was constructed for selected months – May and June for summer and December and January for winter – in 2006 to 2014 (Figure S1). The exponential equation $X = (\log(10.46) - \log(Y))/10.29$ was used to calculate the extinction profile for the trajectory heights, where Y is the vertical height in kilometers and X indicates the extinction against the height of trajectories. Height of individual trajectory points was put in the above equation and got a normalized extinction profile by assuming surface extinction =1(Figure S1).

The WRF-STEM model was used as a third approach for identifying the origin (source regions) of air masses carrying pollutants. Region tagged CO tracer is a standard air quality modeling tool used by other regional and global chemical transport models to identify pollution source regions (Chen et al., 2009; Park et al., 2009; Lamarque and Hess, 2003). The WRF-STEM model uses region tagged carbon monoxide (CO) tracers for many regions in the world to identify geographical areas contributing to observed pollutants (Adhikary et al., 2010). The model domain centered on 50.377° E longitude and 29.917° N latitude. The model horizontal grid resolution was 45x45 km with 200 grids in the east-west direction and 125 north-south. The meteorological variables needed for the chemical transport were derived from the Weather Research and Forecast (WRF) meteorological model (Grell et al., 2005) using FNL data (ds083.2) available from the UCAR website as input data. The main aim of the simulation was to identify the geographic locations contributing to the observed pollutants at the field sites. The HTAP version 2 emission inventory was used in our WRF-STEM modeling. The HTAP version 2 dataset consists of multiple pollutants including black carbon and organic carbon. This emission inventory include major sectors such as energy, industry, transport, residential except large scale open agricultural and open forest fire burning. The simulations applied in our study used the anthropogenic emissions from HTAPv2 inventory (available from http://edgar.jrc.ec.europa.eu/htap_v2/). So the results indicate the

2.5.1 Wind maps, why not use 2005-2006 wind maps instead of 50-year average? 700mb is very high level for low-elevation region and aerosol concentration is very low at that high level. I would suggest use the terrain-oriented level like sigma level near the surface. I would also strongly suggest (a must do) use MERRA-2 reanalysis data, in which not only the data quality is better than NCEP/NCAR but also it includes aerosol data that can be used to compare with the measurement and is more appropriate for looking at the long-distance transport. Suggestion

Response:

Our sampling time period was 2015-2016 so we believe the reviewer wanted us to plot wind maps for 2015-2016.

As per your suggestions, we have used the MERRA-2 reanalysis data at 850mb and replotted the wind maps, during the selected months of 2015-2016 as shown for your reference below. The same figure has been used in the revised manuscript.
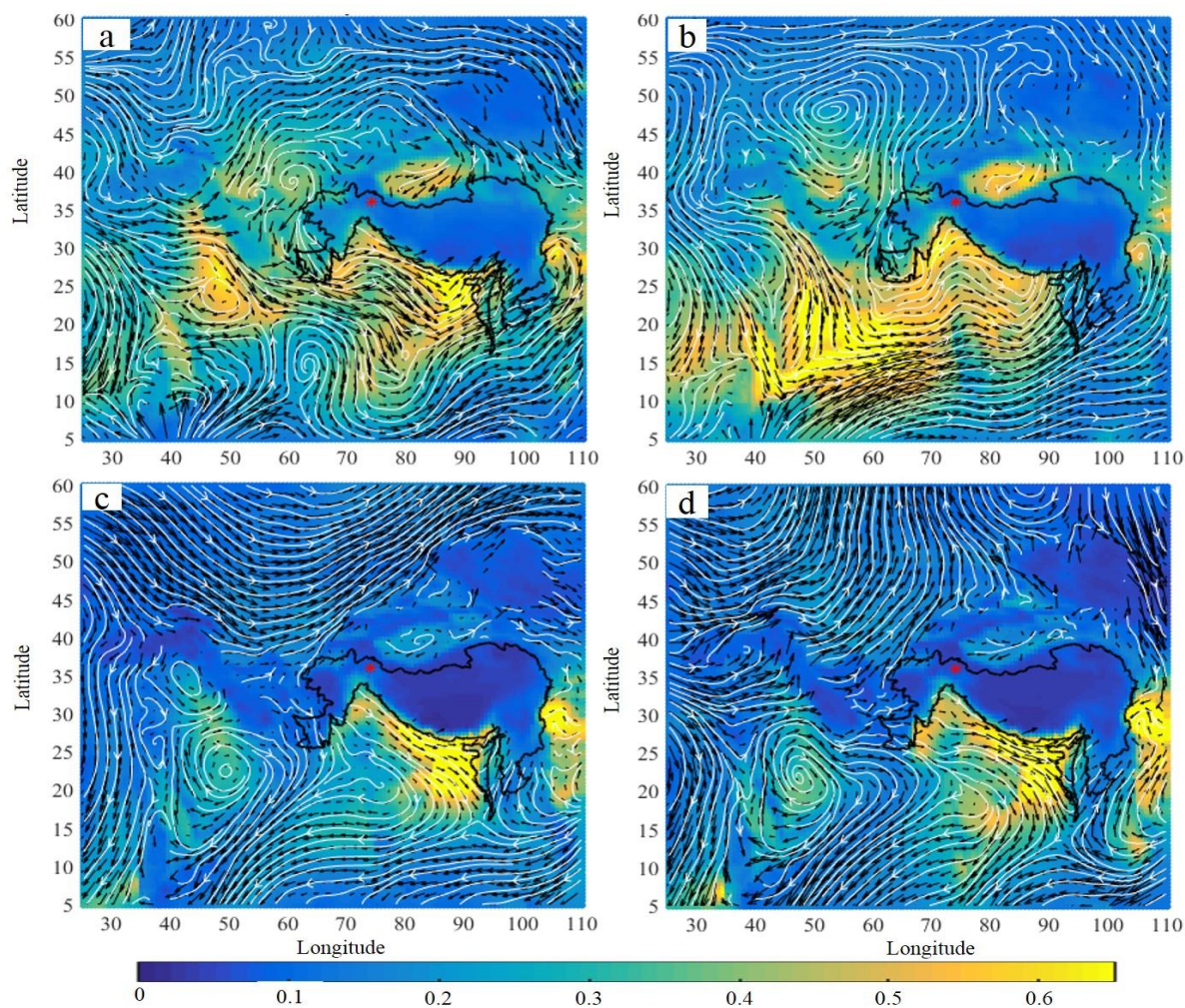
Figure 5. Monthly average horizontal wind patterns at 850 hPa during a) May, b) June, c) December, and d) January, corresponding to approximately 2500 masl, from GES DISC. The study area is indicated by a star, and white lines indicating streamlines. The background colors show monthly mean aerosol optical depth.

2.5.3,

    a. WRF-STEM can only tag CO, because of many differences between CO and LAP in such as emission sources, chemistry and removal, how to quantify their differences in long-range transport and source identifications?

    b. How to infer the transport and source for LAPs based on CO and what's the uncertainty?
    Please see Zhang et al. 2015 and Wang et al., 2015 for source detection methods used over Tibetan Plateau region.

Response:

a. We agree with the reviewer comment, however for the time being we have CO tracer data (which has relatively good correlation with BC tracer in dry seasons) and high resolution BC tracer will be used in our next publication in near future (indicated in lines 587 and 677-680).

➢ BC and CO both are primary pollutants and emit from almost similar anthropogenic sources (may be different in concentration). We were more precise on the source region identification of these pollutants.

➢ Region tagged CO tracer is a standard air quality modeling tool used by other regional and global chemical transport models to identify pollution source regions (Chen et al., 2009; Park et al., 2009; Lamarque and Hess, 2003) lines 249-251. The WRF-STEM model uses region tagged carbon monoxide (CO) tracers for many regions in the world to identify geographical areas contributing to observed pollutants (Adhikary et al., 2010), lines 252.

➢ Each model has some strength and some weaknesses. The strength of our applied model is its relatively higher spatial resolution (line 253). For rough and complex topography of mountain region, it is important to use high resolution model.

b. Transport, source of LAPs based on CO and uncertainty
    Source

        ➢ Depending on event, the source of LAPs (BC) and CO may be different, but in several cases the source of BC and CO might be same such as biomass burning (cooking, outdoor burning, and forest fires) and incomplete combustion process.

        ➢ During incomplete combustion of carbon-containing fuels, such as gasoline, natural gas, oil, coal, and wood, emits CO as well as BC.

    Transport

        ➢ Based on chemical reactivity, the weight of the particle and its life time in the atmosphere, the transport of BC and CO may be different.
        During dry seasons, CO and BC have quite similar transport but in wet seasons (especially during monsoon seasons) BC particles are washed out with precipitation and relatively higher concentrations of CO are reaching receptor

sites as compared to BC. Below quoted text has been added in the revised manuscript (lines 674-675).

"In our case we analyze the model from 1st June to 4th July during summer season and 15th December to 17th January during winter season. In Pakistan monsoon is generally starting during the first week of July each year, so we are expecting relatively good correlation in transport between CO and BC during pre-monsoon period. A cool, dry winter starts from December through February each year."

<u>Uncertainty</u>

Related uncertainty is mentioned in lines 671-680, given below for your reference.

"On modeling side the possible uncertainties are related to using CO as a tracer for light absorbing particles source region. Uncertainties are also attributed to errors in emissions inventories, simulated meteorology and removal processes built in the model. The physics and chemistry of removal for BC and CO are different from each other especially during wet seasons. We analyze the model during pre-monsoon and relatively dry periods, so we are expecting relatively good correlation in transport between CO and BC. While using global emission inventories we were unable to capture emissions at local scale. Contributions of local sources may be underestimated by coarse-resolution models. Therefore high resolution models and emission inventories at local scale are required to capture local emissions. Better-constrained measurements are required in the future for more robust results. High resolution satellite imagery, high resolution models and continuous monitoring can help us to reduce the present uncertainty."

Zhang et al. 2015 and Wang et al., 2015 are presenting impressive work for source detection methods in the Tibetan Plateau region. We have cited these papers in the proper locations.

We agreed with the reviewer comment and using high resolution BC tracer WRF-STEM model in our next publication.

Line 257, 24 hours. Considering–>considered.
Response:
        Corrected (line 301, in the revised manuscript)

Line 318, Jun–>June.

Response:
        Corrected (line 384, in the revised manuscript)

Section 3.2, very weak! How to connect the conclusion from this section with other parts?

Response:

We have rewritten and improved the section according to the suggestions. Repeated sentences have been removed. Some additional text has been added to improve the connection of conclusions from this section to other parts of the paper, lines 379-390.

"The CALIPSO aerosol type identifications analysis indicated that "smoke" was the most frequent-occurring type of aerosol over the study region during both summer and winter seasons. This result indicate that biomass burning sources may be the dominant contributor in this region. Frequency of subtype aerosols for the month of June in 2006 to 2014 is shown in Figure S4. Figure 2 shows the seasonal results for month of May, June (summer) and December, January (winter) in the form of a box plot. During June smoke had the highest frequency (39%), followed by dust (21%), polluted dust (12%), and others (20%) Figure S4. Overall Smoke, dust and or polluted dust were the dominant subtype aerosols in selected months over the study region. This type of aerosol measurement in the atmosphere is important for our current study because it provides observation based data over the study region. Other approaches used (such as modeling) were based on interpolation not observation. Pollutant deposition depends on the concentration of pollutants in the atmosphere, the results are consistent with the high concentration of BC (from smoke) and dust particles in the glacier and snow surface samples."

Section 4 Summary and conclusion, one more section should be added for discussion in uncertainty and possible future direction for both modeling and measurement campaign. For example, how snow aging (snow grain size) and melting water scavenging efficiency (see Qian et al, 2014) affect the conclusions?

Response:
Agreed. We have added one more section for discussion on uncertainty and possible future directions in section 4, lines 660-671, given below for your reference.

"The overall precision in the BC, OC and TC concentrations was estimated considering the analytical precision of concentration measurements and mass contributions from field blanks. Uncertainty of the BC and OC mass concentrations was measured through the standard deviation of the field blanks, experimentally determined analytical uncertainty, and projected uncertainty associated with filter extraction. According to our understanding the major uncertainty in our study was the dust effect on BC/OC measurement. Warming role of OC was also not included in the current research which was low but significant in several regions (Yasunari et al. 2015). Beside this we think snow grain size (snow aging) and snow texture were larger sources of uncertainty in the albedo reduction / radiative forcing calculations. The measured grain size was usually different from the effective optical grain size used in the SNICAR modeling. Snow grain shape was measured with the help of snow card, but was not used in the online SNICAR albedo simulation model and assumed a spherical shape for the snow grains which may slightly affect the results, because albedo of non-spherical grain is higher than the albedo of spherical grains (Dang et al., 2016). On modeling side the possible uncertainties are related to using CO as a tracer for light absorbing particles source region. Uncertainties are also attributed to errors in emissions inventories, simulated meteorology and removal processes built in the model. The physics and chemistry of removal for BC and CO are different from each other especially during wet seasons. In order to reduce uncertainty in source region high resolution BC tracer are required. Better-constrained measurements are required in the future for more robust results. High resolution satellite imagery, high resolution models and continuous monitoring can help us to reduce the present uncertainty."

Figure 2, this is a poor figure and should be re-designed. For example, reduce the y-axis range from 300 to 150. Btw why the numbers for y-axis are 50, 100, 150, 200, 150 (should be 250?)?
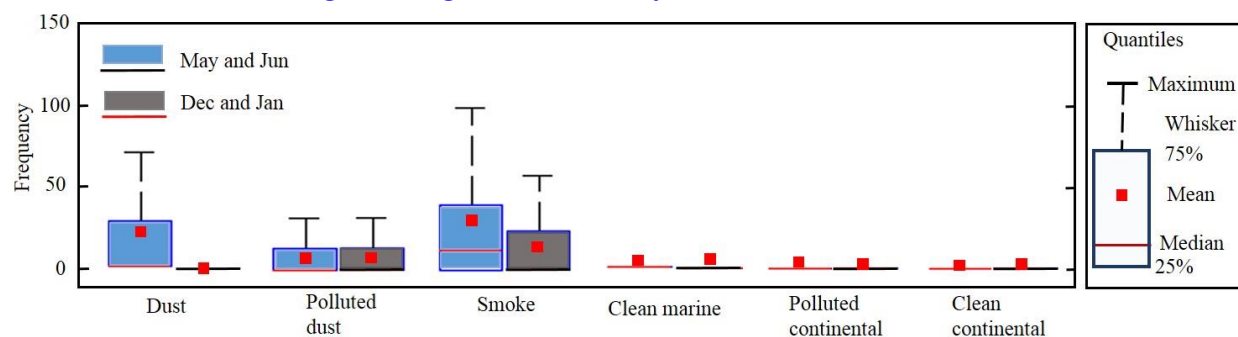
Figure 2. Frequency distribution of aerosol subtypes in the atmosphere over the study region calculated from CALIPSO data for the months in 2006 to 2014.

Figure 3, give full name for MAC in figure caption. Also consider use identical range for y-axis e.g. 0.4-1.0 and for x-axis 0.3-1.2 for Panel c and d.
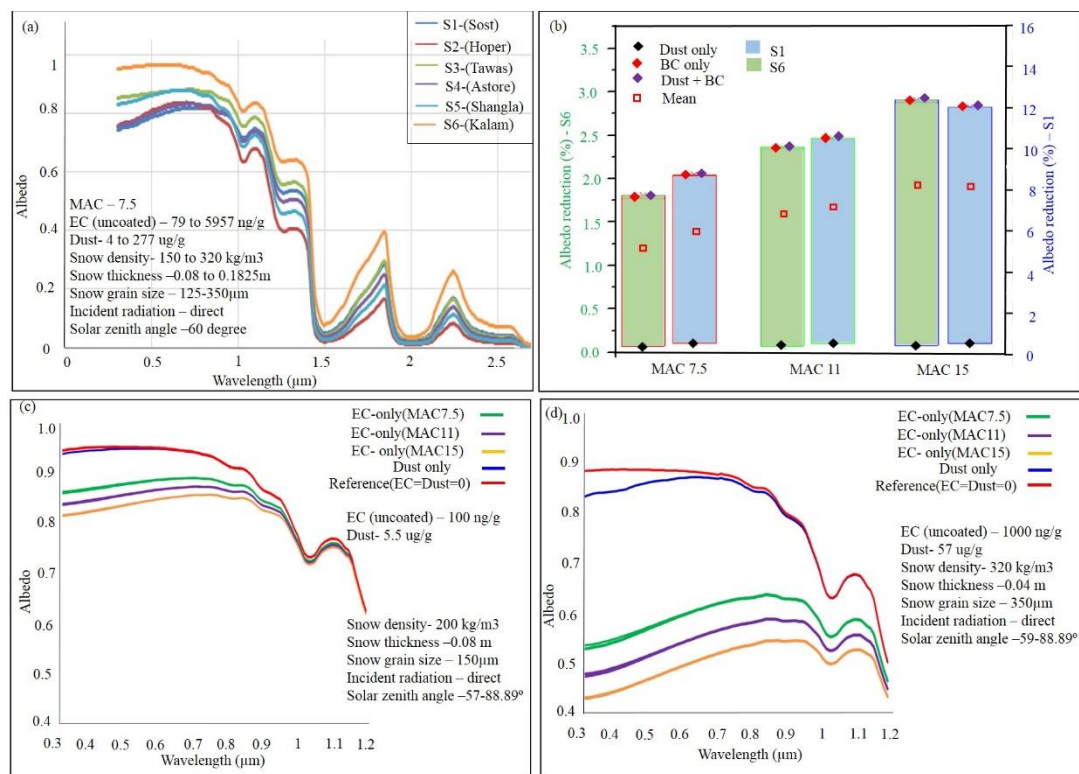
Figure 3. Spectral variation in albedo for winter sampling sites and selected Mass Absorption Cross section (MAC) values, (a) average albedo of samples at each of the sites (b) daily mean albedo reduction of fresh snow (site S6) and aged snow (site S1) snow, (note different scales of y axis) (c) albedo of fresh snow site S6, (d) albedo of aged snow site S1.

Figure 4,

a) **Suggest use identical y-axis range so can highlight the bigger effect over aged snow. The unit for radiative forcing is %?**

b) **More discussion should be provided regarding how snow aging affect the albedo reduction and radiative forcing (e.g. Qian et al., 2014)?**

Response:

a. Agreed. An identical y-axis range has been used. Radiative forcing mentioned here was calculated from albedo reductions indicated on left side of the figure. % symbol has been used in the caption of this figure. Revised Figure 4 is given below for your reference:
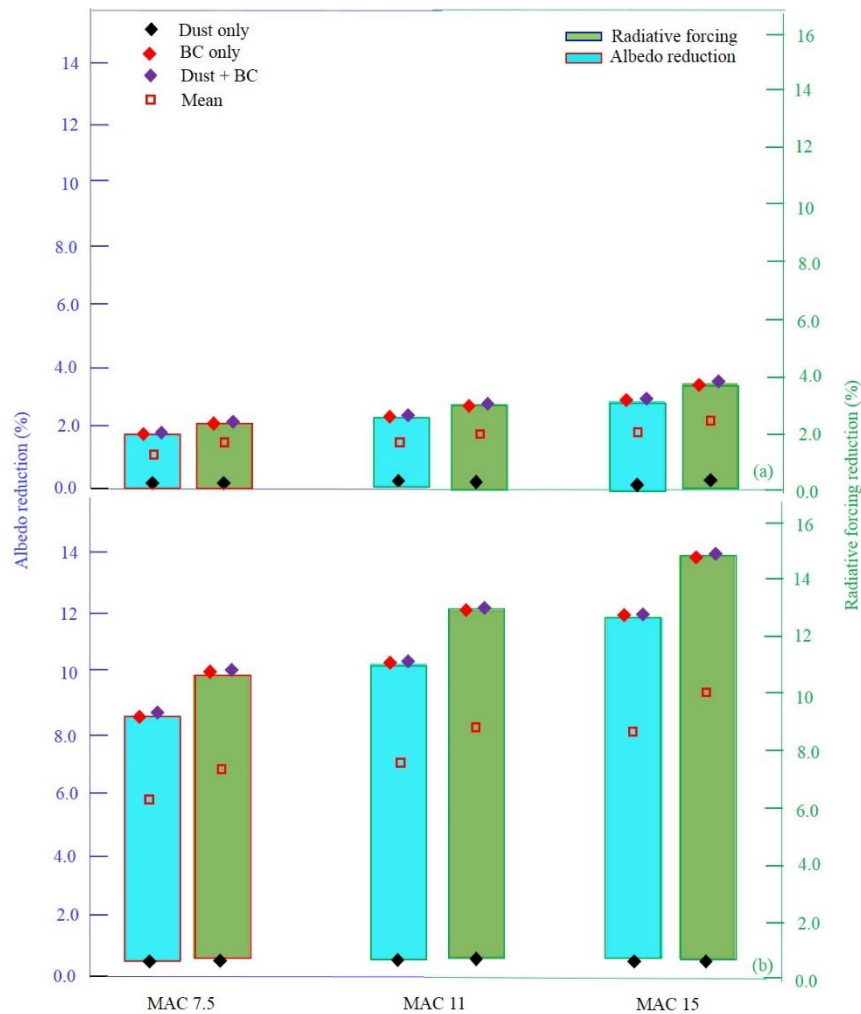
Figure 4. Daily mean radiative forcing reuction and albedo reduction caused by black carbon and dust, for different Mass Absorption Cross section (MAC) in (a) fresh (low black carbon) and (b) aged (high black carbon) snow samples (note different scales of y axis)

b. Agreed. More text has been added regarding how snow aging affects the albedo reduction and radiative forcing (line 505-510), given below for your reference

"Snow aging (snow grain size) plays an important role in albedo reduction and radiative forcing. Schmale et al., (2017) stating that the effect of snow grain size is generally larger than the uncertainty in light absorbing particles which varies with the snow type. The impact of snow aging factor on BC in snow and induced forcing are complex and had spatial and seasonal variation (Qian et al., 2014). Increase of snow aging factor reduces snow albedo and accelerate the snow melting."

Figure 5, what blue contours represent? Again 700 mb is too high and MERRA-2 is a much better dataset.
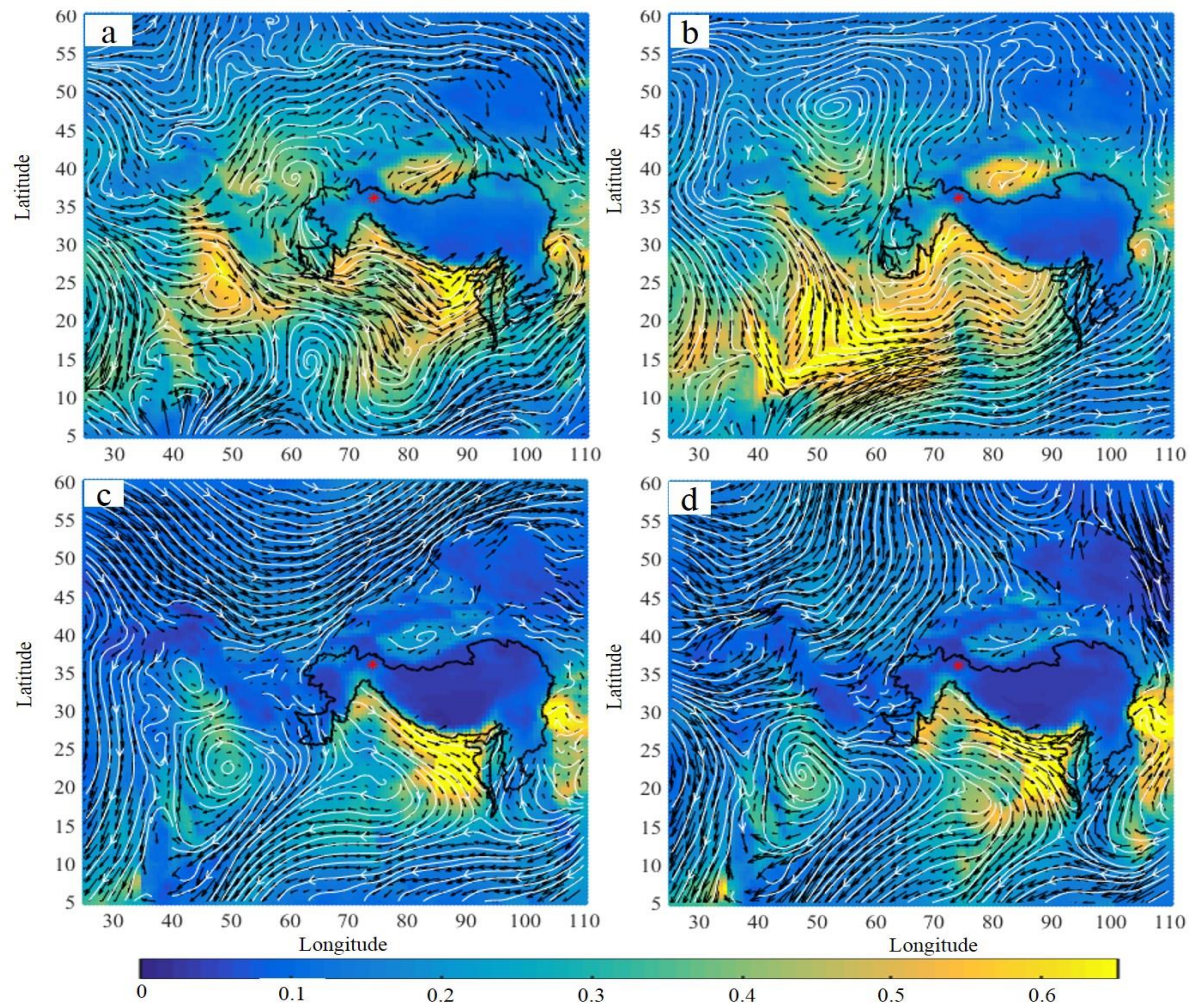
Figure 5. Monthly average horizontal wind patterns at 850 hPa during a) May, b) June, c) December, and d) January, corresponding to approximately 2500 masl, from GES DISC. The study area is indicated by a star, and white lines indicating streamlines. The background colors show monthly mean aerosol optical depth.

Figure 6, not clear what color shades represent?

Possible polluted regions during summer : South west Russia; western part of Kazakhstan, Iran and Turkmenistan; Uzbekistan; Georgia; north east side of Turkey; Syria; Cyprus.

Possible polluted regions during winter : Iran; Pakistan; Iraq; Turkmenistan; Azerbaijan; Georgia; Jordan; Syria; Tunisia; Ukraine; Libya; Egypt; eastern part of Turkey and south western part of Russia.
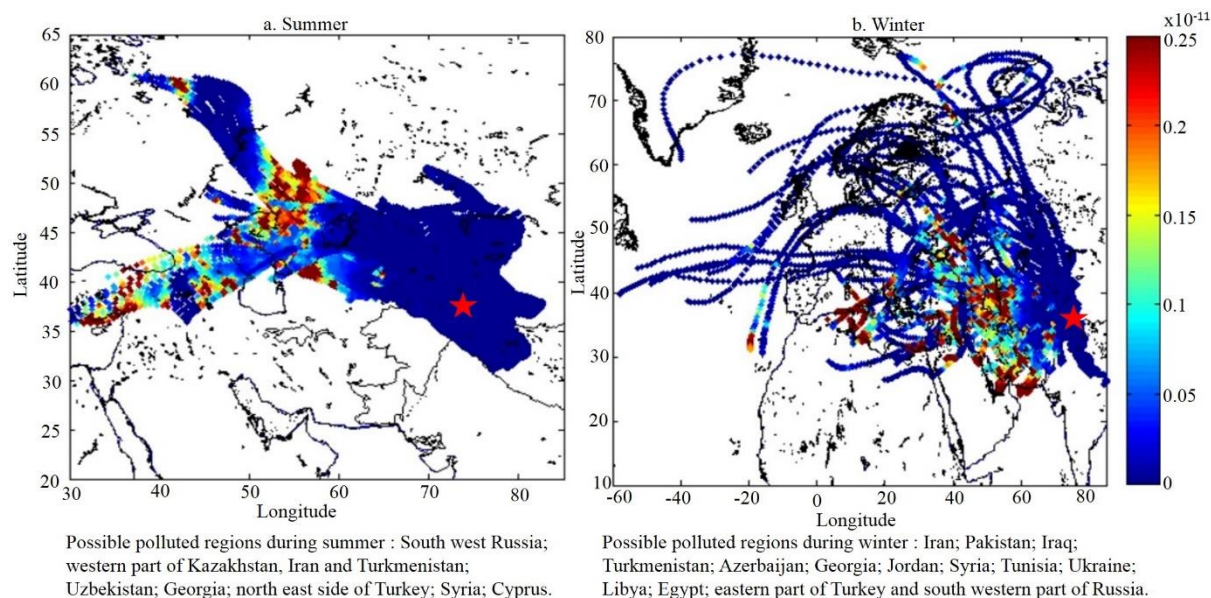
Figure 7,
  (a) I am not sure how the quantitative number of contributions are meaningful because the numbers for LAP could be very different with that for CO.
  (b) Anyway again, Section 2.5 is very poorly organized and kind of just present whatever tools you have or used before, without a clear goal or coherence in science structure. Need to be rewritten with a clear conclusion.

Response:
  (a) Yes, we agreed that the numbers used in the Figure 7 could be different than that for LAP, especially during wet seasons. Based on following justifications, we are expecting relatively less difference between the numbers for LAP and CO.

  ➢ We analyze the model from 1 June to 4 July during the summer season. In Pakistan, monsoon generally starts during the first week of July each year, so we are expecting relatively good correlation in transport between CO and BC during the pre-monsoon period.
  ➢ During winter we analyze the model from 15 December to 17 January. A cool, dry winter starts from December through February each year. Winter season is dry but clouds during this season may bring some uncertainty in our results.

We mentioned this uncertainty in multiple places in the revised manuscript, including lines 672-680. High resolution WRF-STEM BC tagged will be used in our next publication.

  (b) We have reorganized the section 2.5 and made multiple changes. We tried our best to mention a clear goal in this section with a clear conclusion, lines 205-269.

Table 2, give full name for MAC (in other tables/ figures as well).

Response:
Agreed. We used the full name for MAC at all necessary locations, such as lines 48, 58, 61 in supplementary document, and line 917 (caption of Figure 3) in the main manuscript.

Figure S7, give full names for BC1 and BC2.

Response:
Agreed. Full names for BC1 and BC2 has been used, given below for your reference
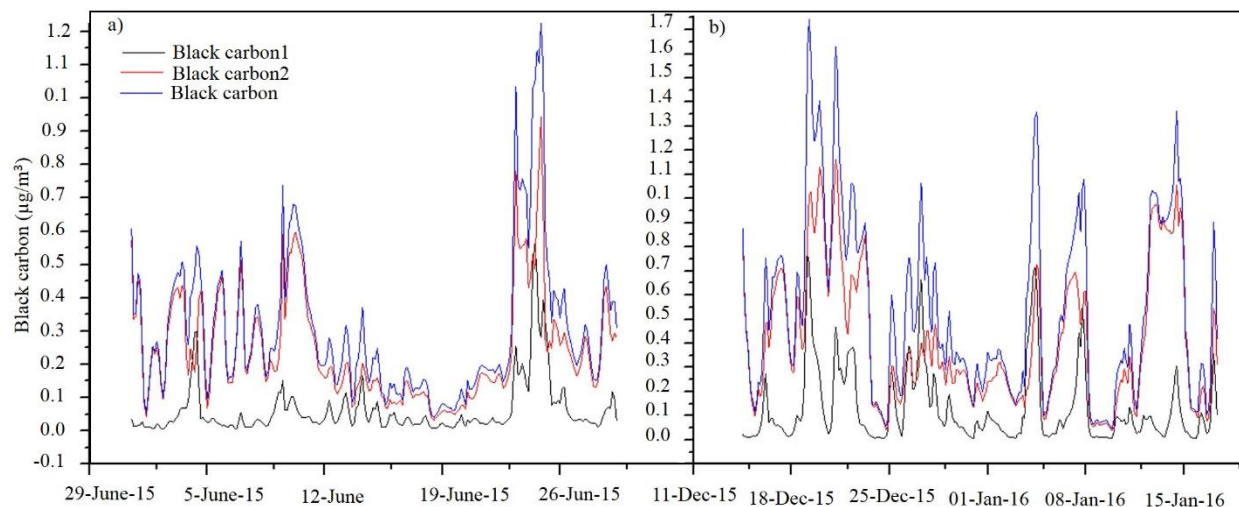

Figure S7. Concentration of black carbon1, black carbon2 and black carbon on the Sachin glacier calculated using the WRF-STEM model: a) summer, b) winter.

Suggested references
  Qian et. al, 2015.
  Zhang et. al, 2015.
  Qian et. al, 2014.
  Wang et. al, 2015.
  Qian et. al, 2011.

Response:
We thank this anonymous reviewer for his careful and detailed reviews and suggestions that helped us to greatly improve this paper. Beside this the provided references/articles are very relevant and presenting a quality work in the region. So we used/cited properly these articles in the revised manuscript. We feel a big improvement in the text after revising and citing these articles.

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Thank you