Anonymous Referee #3, 24 Jan 2022
General

The paper presents BC measurements at a new Arctic site, on the Bely Island in the Kara Sea, Western Siberian Arctic. The origin of the measured EBC and the main contributing sources were assessed using atmospheric transport modelling coupled with the most updated emission inventories for anthropogenic and biomass burning sources of BC.

Bely Island is an important addition to the Arctic network of BC measurement stations because there are not many of them in the vast Siberian Arctic. The fact that it is not far from the oil and gas drilling areas makes it even more important since BC emitted from flaring is a significant climate forcing aerosol in the Arctic.

The comparison of the measured and modeled BC concentrations is excellent. There are many cases when they agree very well. This is important because it shows that the model works. Then there are other cases when there is no agreement at all. This is also a good and important result because it can be used in developing the emission inventories.

The paper is good and I can recommend publishing it in ACP after some minor corrections and additions I suggest below in the detailed comments.

Response: We appreciate reviewer's kind comments and help in improving this manuscript.

Detailed comments

L137 "…Island located in the Kara (Western Siberian Arctic)…" Should it be "Kara Sea", is the word "Sea" missing?

Response: Indeed, this should be "Kara Sea" and we have corrected accordingly. Please see manuscript with Trach Changes page 5.

L137-138 There is a link:
https://peexhq.home.blog/2019/12/11/newresearch-aerosol-stations-in-the-russian-arctic,
When I click it I get this answer
"Oops! That page can't be found. It looks like nothing was found at this location. Maybe try a search?"
Please check the link.

Response: The link that is posted above is misleading. Perhaps this is the reason. We have checked once again and the link that is written in the manuscript (https://peexhq.home.blog/2019/12/11/new-research-aerosol-stations-in-the-russian-arctic) seems to work properly.

Section 2.1 Aerosol station "Island Bely"

I wish you would give some more detailed description of the site and the measurements. After all, this appears to be the first paper on any aerosol measurements at this station so it would make sense to give some more details. I think all the info I suggest you would add would be useful also for future studies at the site.

The site does get heavy snowfall, so was the inlet heated? How was the inlet? Flows? Flow checks?

Response: We have now included a more detailed description. Please see Track Changes page 6: “An aerosol sampling … blocking the system.”

L205-207 "For screening the BC data, we used the measured wind direction. In that case, strong BC spikes that coincided with wind directions related to local diesel sources were removed from further data analyses…wind speed and direction were obtained every 3 hours…"

The AE33 measured at 1-min time resolution and you cleaned them using the wind data at 3-hour resolution. Right?

Response: We have now included more information on how the data screening was performed. Please see manuscript with Track Changes at page 7-8: “Cleaning of 1-min … ” until end of the paragraph.

Did you just assume wind speed and direction remained constant for 3 hours? Ok, if those are the data, that is what can be done. Can you give the contamination sector in degrees? On the other hand, when wind speed is low enough, air will be contaminated regardless of wind direction. That is typical at practically every measurement station. Did you consider this? Note: I don't require you would start reprocessing your data. Can you estimate, how large fraction of data had to be removed due to local contamination?

Response: We did not assume constant winds of course. Wind speed and direction can change within 3 hours. Even in the extreme case where wind blew from the sector 240-250 degrees for 3 consecutive hours (with respect to the meteorological measurements), we did not observe the spike lasting continuously. In such cases, background values are frequent, while the duration of spikes never extends one-two hours even if the wind blew from that sector during a whole day. We have also commented on what happens in low windy conditions. We have now reported the contamination sector in detail and the fraction of data we removed (see manuscript with Track Changes at page 7-8).

Yet another thing is that I think it would make sense to move lines 203 – 208 right after L169.

Response: For the sake of a smoother flow during reading of the current manuscript, we think it would make more sense to present the AE33 instrument and associated parameters first (line 170-203), and then the way we used it at Bely, in order to take the measurements of BC. We leave the paragraph it as it is now, and if Reviewer further insist, we will correct in a future step.

In the small photograph inserted in the map in Fig.1 I see many inlets on the roof of the container. Would you just mention what other aerosol or trace gas measurements you run there? The readers – including myself – could be interested in waiting for followup papers.

Response: We have now added the missing information at page 6, lines 170-175 (please see manuscript with Track Changes).

L233-234 "The source contibution can be displayed as a function of time elapsed since the emission has occurred (i.e., "age")". I am sorry but I don't quite understand this. I

assume "source contribution" means the fraction (in %) of BC observed at the site, coming from some selected source. But the transport time from a source to the station is in hours, both for small and large sources. Please clarify.

Response: We appreciate Reviewer for catching this mistake, because it does not really make sense, as it was written in the manuscript. We have now corrected the whole paragraph. Please see Track Changes at page 8.

L280 3.1 Monthly climatology of black carbon

I have a suggestion: why don't you present the main results also in a table? I am addicted to tables. For future readers and authors it would be very handy to find the results in the table and refer to it, wouldn't it? It is up to you.

Response: Of course! This can be very useful! We have added a new table in Supplementary information for the monthly climatology of BC (see Table S1).

L312-313 "… monthly median EBC in September 2020 demonstrated the unprecedented high value of 72 ng/m3, twice as much as in September 2019."

In Fig 4a, is the black line denoted by "EBC(880)" the median EBC? If so, when I draw a horizontal line from its September 2020 value to the y axis it is clearly lower than 50 ng/m3, not even close to 72 ng/m3. Please check either the data in the figure or the statement above. And by the way, the lines in the legend for EBC(880), quartile25 and quartile75 look just the same. You could use some different line types. And further, please try to avoid plotting data using red and green lines in the same figure, I have colleagues who don't see the difference.

Response: We thank the reviewer for observing this. 72 ng/m3 is mean value for September 2019 and NOT 2020! We have corrected both the value and the Figure to be visible for people who cannot distinguish colors (see Track Changes and updated Fig.4).

L411 " Looking closely to specific episodes, during pollution P1, three events…". Add the word "episode" after "P1"

Response: We have corrected this part everywhere in the manuscript (please see Trach Changes).

About naming the episodes. Now you call them with the same codes P1, P2, … for both the cold and warm seasons. Sometimes this is a bit confucing. In Fig. 5 you show all of them, there are 15 episodes. Consider giving unique numbers for example by simply running numbers P1 … P15 or P1c … P8c and P1w … P7w. Again up to you, but I think it unique names would make it easier to follow.

Response: We have now renamed all episodes as C1, C2, etc… for the Cold period and W1, W2, etc… for the Warm (see updated figures).

L 481 "…"Island Bely" station (Figure 5c)…." There is no Fig 5c, just 5a and 5b. Correct something.

Response: Reviewer has a point here; it should be Figure 5b. We have corrected accordingly. Please see Trach Changes in page 16.

Anonymous Referee #1, 27 Jan 2022
Manuscript No.: ACP-2021-876

General comments:

The Arctic has warmed three times more quickly than the planet as a whole, as the most sensitive area for climate changes. To understand the impacts of BC emissions on the arctic from source regions, particularly from the Siberian Arctic, the authors reported new measurements of equivalent BC (eBC) concentrations for the period of 2019-2020, carried out at the recently established station "Island Bely" which is at the Siberian gateway of the highest anthropogenic pollution to the Russian Arctic.

Through coupling with FLEXPART Lagrangian particle dispersion model and the most updated BC emission inventories for anthropogenic and biomass burning sources, a detailed aerosol aging spectrum, the source region attribution and the source sector apportionment have been investigated for the entire period as well as for the pollution episodes. This is a nice work showing that the observations verified the model simulations and the emission inventories, as well as that the model was able to provide detailed source attributions in terms of emission regions and sectors. Interesting results include

Russian emissions dominate during the entire year, while European and Asian emissions contributed up to 20% in the cold periods;
the annual contribution from anthropogenic sources is dominant, ranging from 75 to 80%;
FLR and BB emissions contribute the largest share of EBC to the "Island Bely" during the cold (by FLG) and warm (by BB) period, respectively;
Gas flaring (FLG) is dominant during cold season (Nov – May) over all the anthropogenic sectors ranging from 47 to 68%;
Biomass mass burning played the biggest role during warm seasons (Jun- Oct.), contributing ~ 80% as the maximum in July;
Those results have improved the source apportionment of Siberian arctic BC, particularly for gas flaring and wildfire impact. This manuscript should be accepted for publication with minor revisions (see the specific comments below).
Response: We appreciate Reviewer's positive manner and his/hers kind comments.

It would be nicer if the contents in sections 3.2, 3.3 and section 4 are presented further succinctly in the revised version.
Response: We have done our best to correct all sections following the very useful comments from the reviewers.

Specific comments:

L37: Based on Table S2, the maximum value of BB is in July instead of June.
Response: We appreciate the reviewer for catching this typo error. It has been corrected in Line 37 (see manuscript with Track Changes).

L38-L39: Based on Table S4, for the BB events during warm seasons, the AEE varies between P2 (BB: ~ 64%) and P6 (BB: > 99%), ranging from 0.8 to 1.35. This

suggests that AAE is not a sensitive tracer for distinguishing BC between anthropogenic and biomass burning sources. This sentence needs to be rephrased.

Response: We have rephrased this sentence stating that AAE during BB events was mostly above 1, excluding two outlier values (see manuscript with Track Changes, Line 39).

L148-L151: This sentence is not well expressed and please re-phrase it.

Response: We have rephrase this sentence as reviewer suggested! Please see L.153-156 in the manuscript with Track Changes).

L222-L224: I am wondering why the authors use 1500 kg/m3 as BC density instead of 2000 kg/m3.

Response: For the BC tracer, we used a density of 1500 kg/m3, which is the one we traditionally use and same as in Stohl et al. (2013). Our choice originates from a very useful review article (Long et al., 2013), who gathered all available measurements for the density of ambient BC. All values are between 1000-1900 kg/m3 (1-1.9 g/cm3, see Table 2, bottom line in Long et al.)

Stohl, A., Klimont, Z., Eckhardt, S., Kupiainen, K., Shevchenko, V. P., Kopeikin, V. M., and Novigatsky, A. N.: Black carbon in the Arctic: the underestimated role of gas flaring and residential combustion emissions, Atmos. Chem. Phys., 13, 8833–8855, https://doi.org/10.5194/acp-13-8833-2013, 2013.

Long, C. M., Nascarella, M. A., and Valberg, P. A.: Carbon black vs. black carbon and other airborne materials containing elemental carbon: Physical and chemical distinctions, Environmental Pollution, 181, 271-286, 2013.

L263-L266: I am wondering whether the authors use two biomass burning emission inventories (GFED v4.1 and CAMS GFAS). Are there any comparison results between the two inventories?

Response: We thought about this comparison, when writing the manuscript. However, we have decided to exclude any comparison of the two products, because it is beyond the scope of this article. In addition, GFED and GFAS products are very different ones, both in terms of methodology and temporal resolution (GFED4 is given monthly, GFAS daily), hence any comparison/criticism would be unfair. In the present case, we found that GFAS gave concentrations closer to our measurements and captured most of the observed peaks. We believe this is because of the high temporal resolution of this product.

L374-L377: I am wondering if it is possible to provide the uncertainties of the model results in Table S1, S2 and S3.

Response: The model uncertainty cannot be really assessed in a way that good give results in a Table. The reason for this is because model uncertainty originates from a number of factors that do not always depend on the parameterization made in the model. For example, to run FLEXPART we need to know the meteorological conditions at the time of the simulation. This is taken from operational analyses for the European Centre for Medium Range Weather Forecasts (ECMWF). As a separate product, the u and v component of the winds that are necessary to run the model are associated with an uncertainty. Furthermore, other parameters are also used from ECMWF as an input to FLEXPART, which affect scavenging and removal of species from the atmosphere. For instance, the real position of the clouds is also used from

ECMWF, which affect where in-cloud and below-cloud scavenging occurs (see Pisso et al., 2019, of the manuscript), as is again associated with an uncertainty. More than this, the parameterizations that have been performed in the model are associated with an uncertainty (e.g., turbulence). To calculate model uncertainty, separate sensitivity tests have been performed each time investigating how each parameter affects the overall result. Besides, FLEXPART – as an open access algorithm – is of the most widely used algorithms in the world and is continuously validated against observations, while assessing its sensitivity to different model parameters. A few papers that have investigated model uncertainty are the following:

Evangeliou, N., Hamburger, T., Cozic, A., Balkanski, Y., and Stohl, A.: Inverse modeling of the Chernobyl source term using atmospheric concentration and deposition measurements, Atmos. Chem. Phys., 17, 8805–8824, https://doi.org/10.5194/acp-17-8805-2017, 2017.

Evangeliou, N., Thompson, R. L., Eckhardt, S., & Stohl, A. (2018), "Top-down estimates of black carbon emissions at high latitudes using an atmospheric transport model and a Bayesian inversion framework", Atmospheric Chemistry & Physics, 18, 15307-15327, https://doi.org/10.5194/acp-18-15307-2018, 2018.

Kristiansen, N. I., Stohl, A., Olivié, D. J. L., Croft, B., Søvde, O. A., Klein, H., Christoudias, T., Kunkel, D., Leadbetter, S. J., Lee, Y. H., Zhang, K., Tsigaridis, K., Bergman, T., Evangeliou, N., Wang, H., Ma, P.-L., Easter, R. C., Rasch, P. J., Liu, X., Pitari, G., Di Genova, G., Zhao, S. Y., Balkanski, Y., Bauer, S. E., Faluvegi, G. S., Kokkola, H., Martin, R. V., Pierce, J. R., Schulz, M., Shindell, D., Tost, H., & Zhang, H. (2016), "Evaluation of observed and modelled aerosol lifetimes using radioactive tracers of opportunity and an ensemble of 19 global models", Atmosheric Chemistry & Physics, 16, 3525-3561, doi:10.5194/acp-16-3525-2016.

Grythe, H., Kristiansen, N. I., Groot Zwaaftink, C. D., Eckhardt, S., Ström, J., Tunved, P., 673 Krejci, R., and Stohl, A.: A new aerosol wet removal scheme for the Lagrangian particle model 674 FLEXPART v10, Geosci. Model Dev., 10, 1447-1466, 10.5194/gmd-10-1447-2017, 2017.

Pisso, I., Sollum, E., Grythe, H., Kristiansen, N. I., Cassiani, M., Eckhardt, S., Arnold, D., Morton, D., Thompson, R. L., Groot Zwaaftink, C. D., Evangeliou, N., Sodemann, H., Haimberger, L., Henne, S., Brunner, D., Burkhart, J. F., Fouilloux, A., Brioude, J., Philipp, A., Seibert, P., and Stohl, A.: The Lagrangian particle dispersion model FLEXPART version 10.4, Geosci. Model Dev., 12, 4955–4997, https://doi.org/10.5194/gmd-12-4955-2019, 2019

L396-L397: It is not convinced that the observed AAE values in this study are sensitive to BB influenced in both cold and warm seasons (Table S4).

Response: The reviewer is correct here, and this is what we have tried to clarify in this paragraph. Of course, one cannot expect that during a long-range transport event where mixing and aging occur, an AAE value representative for BB will always be observed. However, AAE can be used as a proxy for whether BC originates from biomass burning or fossil fuels. We have tried to rephrase the paragraph (see Lines 464-471, manuscript with Track Changes).

L405-L406: How about the results by FLEXPART with GFED v4.1?

Response: Like we wrote in a previous comment, we do not show he results of the two products to avoid comparisons of two very different fire products (both in

methodology and temporal resolution). We present the closest-to-observations model results we got.

L408-L410: Should the “Figure 5b” be replaced by “Figure 4c” for monthly median contribution of sources to BC in the cold period?

Response: Yes, Figure 4c is the correct figure when it comes to monthly median. We thank the reviewer for seeing this detail. We have corrected accordingly (see Track Changes at page 14).

L525: Typo (?): please replace “ageing” with “aging”.

Response: Not sure what is correct here, but we guess the editorial office will correct in a later stage. According to grammar.com (https://www.grammar.com/ageing_vs._aging), “ageing” is used in British English, “aging” in American English. We used the British version throughput the manuscript and have now changed to American English as reviewer suggested.

L543-L544: This sentence needs to be rewritten. It has been observed that while the AAE value is between 1- 1.35, the BC could be also influenced dominantly by FLG (Table S4).

Response: Very good point! We have corrected this part as reviewer suggested. Please see Line 662 in manuscript with Track Changes.

L862-L874: Is it possible to have the data plotted in Figure 4b, 4d included in individual Tables of the supplement as the Table S1 for Figure 4c? Each of the Tables should also include corresponding AAE values.

Response: The monthly climatology of BC is now given in Supplementary Table S 1. In addition, we have made all the model results from this study publicly available, including plots and ascii files with source, continental contributions, ageclasses etc… Please find all the results in our interactive webpage (https://niflheim.nilu.no/NikolaosPY/Bely_2020_cams.py). This is also highlighted in the Data availability statements at the very end of the manuscript.

L892: At the end of line, the "(bottom row)” is missing.

Response: Legend in Fig. 7 was corrected. Same correction was performed in the Legend of Fig. 8 (manuscript with Track Changes).