<u>Reviewer 1</u>

We thank the Reviewer for her/his detailed comment. We address all her comments below, where highlighted text is the Reviewer's comment followed by the unformatted text of our reply.

In this case I am rather satisfied with the revision. There is still confusion regarding the application of the model to beryllium isotopes and then its presentation and validation considering just Be-7. Also, I do not completely understand the connection between a beryllium model with eventual application to Be-10 data with the choice of high-latitude regions and annual timescales. Could you please better specify this?

As explained in the revised introduction, our eventual goal is to understand the production/transport of 10Be measured in polar ice cores with annual resolution. 10Be is a primary isotope to study cosmic-ray and SEP variability on the long-term scale before the era of direct measurements (e.g., Beer et al., 2012; Usoskin, 2017). Unfortunately, the existing models are unable to fully reproduce the measured data in polar ice, and higher accuracy of models is needed. This is why we are primarily interested in polar regions and annual time scales. The purpose of this paper is to check and validate the ability of the model to perform the first step, viz. production and transport of beryllium. However, the model output can hardly be compared directly to data because 10 Be is not measured in the air but rather as 10Be concentration in the ice where it is additionally affected by the deposition and possible post-depositional effects. These effects are left for forthcoming studies and not touched here. The only way to validate a beryllium transport model is to use 7Be which is routinely measured in air samples. 7Be is similar to 10Be in production and transport but has a different lifetime which is easy to account for. Thus, we compared the model output for 7Be with the measured near-ground air concentrations focussing upon high-latitude regions and annual timescale, as explained above. We are happy that Reviewers 2 and 3 understand our motivation. As Reviewer 2 states "And yes, there are many papers regarding the 7Be in mid latitudes, but these data cannot be used and/or applied to 7Be at high latitudes and its behavior (similarly 10Be). And only in Polar Regions can be studied the solar energetic-particle event (SPE) and their influence in 7Be concentrations."

As commented below in the specific comments, there are still some deficiencies in the added Section 2. Indeed, the review of earlier models is far from being complete, missing important works on this subject. It is true that this field is still widely open for improvements, but the state-of-the-art in this research field has to be consistently described. Secondly, to me the newly added Section is part of the Introduction section, being fundamental for the reader to understand the need of this particular work and its scope. Therefore, to be the introduction of this as Section 2 generates confusion and does not fully help to address the above reported issues.

We disagree with this comment. Moving discussion of previous models to the Introduction would make it heavily distracting the reader's attention from the formulation of the aims of the paper. Instead, a separate focused Section 2 makes it easier to read.

As commented below, the histograms shown in Figure 11 actually indicate that apart from the mean value of the bias, there are many cases, especially in southern stations, when the bias is actually higher and close to 1. Indeed, the mean low value could result from the distribution of the bias. In addition, the histogram may not be the best way to represent the bias, since the shape of the histogram is dominated by the choice of the bin width. As I previously mentioned, there are a set of statistical parameters to calculate and validate a model. In addition, it would be interesting to evaluate the pattern of the bias to investigate if it is higher during particular seasons or if conversely the high biases are randomly distributed.

We believe that this comment is caused by confusion. Figure 11 shows that there is no significant bias between the model results and measurements. The Reviewer apparently confuses the *bias* which is a systematic displacement and random *deviations*. The histograms imply that there are no biases but deviations

can be large. However, these discrepancies are not worrisome for our purpose because they occur on a synoptic timescale which is known to be not reproduced by the general circulation models (e.g., Usoskin et al., 2009, Brattich et al., 2017). Deviations are larger during the local summer season, in agreement with other models (Brattich et al., 2017). As one can see from Fig.10, the agreement at the annual scale is always good. The exact shape of the histogram may indeed depend on the bin width, but the metrics of the distribution (the mean and standard deviation) are defined unambiguously by the dataset and do NOT depend on the parameters of the histogram.

In our view, the presently used standard set of statistical metrics is sufficient to validate our model: the correlation/coherence analysis confirms that the modelled and measured variabilities of 7Be concentrations agree very well on the annual scale, while the distributions of the residuals confirm the absence of any significant bias between the model and the data.

Ok with this addition. However, it is still not clear which kind of meteorological data have been used.

CCM SOCOL uses ECHAM5 nudged with ERA-Interim (eraiaT42L39) reanalyses. Full nudging is a linear relaxation of thermodynamic parameters: temperature, divergence and vorticity of the wind field.

It is true that large-scale dynamics is strongly different in the different seasons, which on average can perhaps be described as you did above, even though there can be situations largely deviating from this average behaviour. In addition, the situation that you depict is not the one that is observed in reality and discussed in many scientific papers in the field. It is in fact well-known that even though the rise of the tropopause and the presence of intense convection during the warmest seasons favor the transport of upper tropospheric air to the surface, the dynamics and the instability of the coldest seasons tend to favor intense intrusions from the stratosphere to around. However, these events of stratospheric intrusions and transport of air from the upper troposphere-lower stratosphere have to be rather intense to be observed at ground, and are usually observed only at high-altitude stations. In addition, these events do not represent the average situation, but rather a particular one extending for two or three days. Finally, even though these events are typical of the cold season, they sometimes occur even during the other season. There are lots of papers describing the dynamic of stratosphere-to-troposphere transport, a topic that though still not completely understood is not completely obscure and subject of many scientific projects/papers. Just to cite a few of the most important ones, you can see for instance: Cristofanelli et al., 2003. Stratosphere-to-troposphere transport: A model and method evaluation. Doi:10.1029/2002JD002274; Zanis et al., 2003. An estimate of the impact of stratosphere-to-troposphere transport (STT) on the lower free tropospheric ozone over the Alps using 10Be and 7Be measurements. doi:10.1029/2002JD002604; Stohl et al., 2003. Stratosphere-troposphere exchange: a review, and what we have learned from STACCATO. Doi:10.1029/2002JD002490; Tarasick et al., 2019. Quantifying stratosphere-troposphere transport of ozone using balloon-borne ozonesondes, radar windprofilers and trajectory models. Doi:10.1016/j.atmosenv.2018.10.040.

My comment was aimed at clarifying exactly that if you do not present the synoptic situation characterizing the SEP event and the other seasons, you cannot be sure that the beryllium produced in the upper atmospheric layers is then transported to ground and therefore your analysis of the event during the SEP dates and in other dates in other seasons likely could be not capable of representing the effect of seasonality on beryllium concentrations.

An analysis of the synoptic situations would be definitely of interest, but it lies beyond the scope of this manuscript, which aims at the description and evaluation of the new model, as explained above, with focus upon high-latitude regions and annual timescale. For our case, the representation of more robust systematic seasonal STE changes is more important.

Specific comments

1) Line 35: Change "from" to "form".

Done.

 Line 40: Delete the first comma. Change "cadences" to "time resolution" or "sampling resolution" or similar. Consider revise the sentence which is quite long and not totally clear.

It is changed to "time resolution"

3) Line 45 and throughout the manuscript: Again change the term "cadence".

It is changed to "time resolution".

4) Lines 4-6 and 48-49: In general the models do not reproduce only the transport of beryllium isotopes, but also at least their deposition, and a parameterization of sources (maybe not as accurate as yours), and thus the simulated concentrations do not depend only on transport. This further means that in most cases, input meteorological fields should contain not only wind data. Please consider revise this.

Lines 4-6: While transport of ⁷Be can be modelled with high accuracy using the known meteorological (wind) fields, atmospheric transport of ¹⁰Be was typically modelled using case-study specific simulations or simplified box models based on parametrizations.

We rephrased it as

Lines 5-7: While transport and deposition of ⁷Be can be modelled with high accuracy using the known meteorological fields, ¹⁰Be was typically modelled using case-study specific simulations or simplified box models based on parametrizations.

Line 49: Its atmospheric transport can be modelled with high accuracy using the known meteorological (wind) fields.

We rephrased it as

Line 49-50: Its atmospheric transport can be modelled with high accuracy using the known meteorological fields.

5) Lines 49-50: Please check the two verbs, one is singular and the other is plural.

Done, see point 4.

6) Lines 56-58: A reference is needed for this sentence.

The following reference has been added to the list:

U. Heikkilä, J. Beer, J. Feichter. Modeling cosmogenic radionuclides 10Be and 7Be during the Maunder Minimum using the ECHAM5-HAM General Circulation Model. Atmospheric Chemistry and Physics, European Geosciences Union, 2008, 8 (10), pp.2797-2809. ffhal-00296559

7) Lines 58-60: This sentence, which introduces the scope of this work, is connected with the sentences at lines 107-109, 116-117 and further. I understand the need to introduce the new Section 2 for presenting the state-of-the-art models on this topic, but then wouldn't be Section 2 a part of the Introduction, and thus better presented as Section 1.1? This revision would need not only a change in the numbering, but also a better formulation of the scope of the article and its structure after presenting the gaps of past and recent works.

We do not agree with this suggestion (see above).

8) Line 82: Change "ModelE" to "model".

The term "ModelE" is correct, see description at https://www.giss.nasa.gov/tools/modelE/

9) Lines 70-94: The review of earlier works is not complete. For instance, much of the work conducted by NASA with the GMI and GEOS-Chem CTMs is missing. Please consider revise conducting a careful review including a more complete overview of the modeling work to represent beryllium isotopes.

We have added information about GMI CTM in the review of earlier works. However, the CTM approach applies only to years where the meteorological fields are known, while CCM can work self-consistently without prescribed meteorology. Accordingly, CTM models cannot be used to study beryllium isotopes (mainly, ¹⁰Be) in the past or future scenarios, that is the main aim of this work.

10) Line 99: The knowledge of the meteorological fields should be essential to all modeling simulations of this kind, so I do not see this as a significant gap of this methodology.

We are talking about prescribed observation-based meteorology, which does not exist for the past and future, while our primary goal is to model ¹⁰Be production/transport/deposition for the past data and future-projection scenarios.

We rephrased line 127 as:

The latter approach is applicable only to recent years where the observed meteorology is known.

Moreover, this approach does not include either stratospheric dynamics or depositional processes and, therefore, is not suitable for ¹⁰Be in polar ice.

11) Line 104: Delete comma.

Done.

12) Lines 104-106: I cannot fully understand those requirements: do you mean that in general, the climatological and beryllium communities do not collaborate?

Unfortunately, yes. The collaboration ought to be much closer.

13) Lines 175-176: Which data? Of which variables?

Full nudging means a linear relaxation of thermodynamic parameters: temperature, divergence and vorticity of the wind field.

14) Line 229: Change to "with smaller absolute values".

Done.

15) Lines 291-296: References are needed.

The following reference has been added:

Terzi, L., Wotawa, G., Schoeppner, M. et al. Radioisotopes demonstrate changes in global atmospheric circulation possibly caused by global warming. Sci Rep 10, 10695 (2020). <u>https://doi.org/10.1038/s41598-020-66541-5</u>.

16) Figure 11: If the caption is correct and the histograms reproduce the difference between modeled and observed 7Be values, then the title of the x-axis is not. Although the mean reported values are indicative of a low bias, the figure shows that there are cases in which the bias is high, almost equal to 1 in Chile and at Kerguelen, thus contradicting some statements by the authors. In addition, since it is known that the shape of histograms is dominated by the correct choice of the bin width, couldn't you think of a different method to represent the bias?

We don't understand why the Reviewer believes that the title of the x-axis is not correct. The Figure caption clearly says that this is the difference between modelled and measured 7Be weekly activities, and the unit of

the difference is indeed the activity, viz. mBq/m3, as written in the X-axis title. As discussed above, the Reviewer apparently confuses concepts of "bias" (systematic discrepancy) and "deviation" (instant difference). This Figure shows that there is no significant/notable bias between the model results and the data, but the deviations can be indeed large on the synoptic (weekly) timescale. However, since we are mainly interested in the annual timescale, this is not a problem. While the shape of the histogram may slightly depend on the bin width, the parameters of the distribution of the residuals (the mean and the standard deviation) are unambiguously defined by the data and have nothing to do with the histogram bin width.

17) Line 348: The paper of Brattich et al. (2021) has not been added to the reference list.

The paper of Brattich et al. (2021) was added.

18) Revise the acronym as being SPE and not SEP throughout the manuscript and figures. It is fixed as SPE throughout the paper.

Reviewer 2

We are thankful to this Reviewer for her thorough and in-depth comments, deep knowledge of the field and positive attitude.

Proposed revisions:

-1-

In my opinion, there is a confusion with the names-abbreviations of the models that are mentioned in this work (*SOCOL-AERv2-BEv1, SOCOL, CCM SOCOL, SOCOL v3.0, SOCOL-AERv2, CCM SOCOLAERv2, SOCOLv3.0:Be, SOCOLv3.0*). See abbreviations in text at section -4- bellow.

My suggestion to the authors is to try to "keep a unique form" as much as possible with the model abbreviations, e.g. I do not understand if there is any difference between the CCM SOCOL and SOCOL. For sure if there is a difference they authors should keep the different abbreviations, but if not please keep just one.

<mark>-2-</mark>

The name of the model that is developed here **SOCOL-AERv2-BEv1**, is mentioned only once in the title and once at the line 123 of the manuscript.

In my opinion, the name of the model must be added in the Abstract and in the Conclusions. -3-

Furthermore, since it is "*a new developed model*" it could be mentioned somehow in the title. But this is not mandatory and the authors will decide for the title of the final paper. It's just a proposal. -4-

Below I mention some parts of the text with the abbreviations of the model that at least in my case produced a slight confusion.

Title: ".....SOCOL-AERv2-BEv1...."

<mark>Abstract</mark>

Lines 12-13: "based on the chemistry-climate model SOCOL (SOlar Climate Ozone Links) v3,…" lines 25-26: "Thus, a new full 3D time-dependent model, based on the SOCOL v3.0, of 7Be and 10Be atmospheric production, transport and deposition have been developed."

In my opinion, the development of the new model must be mentioned in title.

<mark>1 Introduction</mark>

Lines 58-60: "Here we develop such a model to trace isotopes of 7Be and 10Be in the atmosphere

based on the **chemistry-climate model SOCOL (SOlar Climate Ozone Links) v3**, which has been specifically modified by including modules for the production, deposition, and transport of 7Be and 10Be."

2 Summary previous and existing models

Lines 108-110: "Here we present a new development of the **full chemistry-climate model (CCM) SOlar Climate Ozone Links (SOCOL)** for modelling of production, transport, and deposition of the cosmogenic isotopes of beryllium as well as its validation with the available measurements of 7Be at high-latitude locations. The **CCM SOCOL** is potentially capable of simulation"

Lines 112-115: "A recent model version **SOCOL-AERv2**, which simulates aerosols more realistically does not, however, include the treatment of all processes relevant to the beryllium life cycle and its applicability has not been evaluated. We have **further upgraded the CCM SOCOL-AERv2** (Feinberg et al., 2019) here, by adding the production, transport, and deposition of 7Be and 10Be isotopes from both GCR and SEPs."

Lines 123-124: "The new model version **SOCOL-AERv2-BEv1** has been developed here for systematic modelling of 7Be and 10Be production, transport and deposition in the atmosphere."

It is the first time that the authors mention the name of the model that has been developed and presented in this work.

In my opinion, the name of the model must be added in the Abstract and in the Conclusions. 4 Model description

Lines 171-172: "We used an extended version of the **CCM SOCOLv3** (Stenke et al., 2013) with the aerosol module - **SOCOL-AERv2** (Feinberg et al., 2019)."

Lines 179: "SOCOL uses the horizontal resolution T42, where...."

Line 200: "... used as an input for the **SOCOL** model,..."

Line 258: "...are realistically modelled by the CCM SOCOL (Feinberg et al., 2019)...."

6 Evaluation of the model by comparison with direct 7Be measurements

Line 387: "...deposition modelled by SOCOL..."

Conclusions

Line 394: "...The model named as **SOCOLv3.0:Be** is based on the chemistry-climate model **SOCOL,** specifically.."

Line 411: "Concluding, a new full 3D time-dependent model, based on **SOCOL-AERv2**, of 7Be and 10Be...."

We thank this Reviewer for her valuable suggestion. We now use only two terms "CCM SOCOL" for the basic version of the model, and CCM "SOCOL-AERv2-BE" for the specific modified version.

Reviewer 3

We thank the Reviewer for his/her useful comments and deep knowledge. We address all the specific comments below.

P11 L225-226: "It should be noted that we treat 7Be as gas only for the advective transport when the result is the same for the small particles and gas components. "- Why the 7Be is treated as gas not the aerosols when the result is the same. The 7Be is treated as the aerosols in both the previous model and the observation. Why is it not sufficient to treat the 7Be as an aerosol?

We agree that the previous formulation was confusing and potentially misleading. In the model, gases and small aerosol particles are transported as passive tracers. Thus, although beryllium is indeed attached to aerosols and considered so in the model, its transport is modelled in a way similar to gases, only for the advective transport. In order to avoid further confusion, we have re-written the text in several places:

(line 271)

It should be noted that we treat ⁷Be as a passive tracer only for the advective transport when the result is the same for the small particles and gas components.

Line (274)

In this study, the advective transport of 7Be and 10Be as passive tracers was performed using Flux-Form Semi-Lagrangian Transport Schemes (Lin and Rood, 1996) embedded in ECHAM5.

Line (288)

It means that beryllium atoms are considered as passive tracer only for the transport process, while with respect to the dry and wet depositions, they are treated as sulfates.

Line (296)

Scavenging coefficients for all tracers are calculated based on Henry's law equilibrium constants.

P12 Figure 4: Isn't the vertical axis the amount of cosmogenic Be isotopes in the atmosphere? I have never seen the concentration of the cosmogenic isotopes as 108 atoms m-3, also in the stratosphere (Jordan et al., 2003).

We agree that the plot and the units were confusing. It has been redone and now shows the globally averaged columnar content of beryllium as the number of atoms per cm2 of the Earth's surface.

P13 L290: Since volume of the air is extremely changing with atmospheric pressure, it is recommended to use at / m3 SPT as the volume unit.

We have reploted fig. 4 using atoms per cm^2 of the Earth's surface.

P15 Figure 6: Thea color contour is unclear to represent the deposition distribution described in the text. It should be made clear that the deposition at the West of the continents is lower than other areas.

It has been added in lines 317-319.

P15 L316: "on the 30th day after the event "- It might be miss touch "on the 30 days after the event."

Done.

P15 L324: Miyake et al., 2018 (GRL https://doi.org/10.1029/2018GL080475) detected the SEP signal at 993-994CE of 10Be in the quasi-annual Antarctic ice core record. Is it possible that the SEP event I stronger than a few orders of magnitude?

Indeed, several events have been detected in the past, particularly in annual 10Be ice-core data. The strongest event of 774-775 CE was a factor ~300 greater than the event of 20-Jan-2005 considered here. We modelled the production and transport of beryllium for a major solar energetic-particle event of 20-

Jan-2005, which may serve as a reference event for historically known extreme SPEs. We have updated the text accordingly.

P16 L329: Figure 9 is not atmospheric concentrations. Please check the data.

We have checked our plot and found it consistent with similar results shown, e.g., in Heikkilä, U. and Smith, A. M.: Influence of model resolution on the atmospheric transport of 10Be, Atmos. Chem. Phys., 12, 10601–10612, https://doi.org/10.5194/acp-12-10601-2012, 2012 (see fig. 8 for L39 therein). The latter is provided only for GCR, while our model includes also SPE, thus, the values in the upper atmosphere are high.

P22 L446: "Three-dimensional simulation of 7be in a global climate model. "- "Three-dimensional

simulation of 7Be in a global climate model. "

Done.

P22 L465: "Modeling production and climate-related impacts on 10be concentration in ice cores. "-

"Modeling production and climate-related impacts on 10Be concentration in ice cores. "

Done.

P23 L500: "Sulfur, sea salt and radionuclide aerosols in giss modele. "- "Sulfur, sea salt and

radionuclide aerosols in GISS modelE."

Done.

P23 L507: "Stratosphere-troposphere exchange in a changing climate simulated with the general

circulation model maecham4. "- "Stratosphere-troposphere exchange in a changing climate simulated

with the general circulation model MAECHAM4. "'

Done

P23 L508: "Deposition of naturally occurring 7Be and 210Pb in Northern Finland. "- "Deposition of naturally occurring 7Be and 210Pb in Northern Finland. "

Done

P24 L513:" Geomagnetic and atmospheric effects upon the cosmogenic 10be observed in polar ice. "-"Geomagnetic and atmospheric effects upon the cosmogenic 10Be observed in polar ice. "

Done.

P24 L522: "Extended versions of the convective parametrization scheme at ecmwf and their impact on the mean and transient activity of the model in the tropics"- "Extended versions of the convective parametrization scheme at ECMWF and their impact on the mean and transient activity of the model in the tropics"

Done.

P24 L525: "Geophysical Research Letters" is not an abbreviation.

Done.

P35 L558: "Global cloud and precipitation chemistry and wet deposition: tropospheric model simulations with echam5/messy1. Atmospheric Chemistry and Physics "- "Global cloud and precipitation chemistry and wet deposition: tropospheric model simulations with EHCAM5/MESSy1. ". "Atmospheric Chemistry and Physics" is not an abbreviation.

Done.

L26 L597: "atmospheric 7Be in Europe"- "atmospheric 7Be in Europe"

Done