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Ref: No.: acp-2022-282

Title: Detection of Stratospheric Air Intrusion Events From Ground-based High-resolution  $^{10}\text{Be}/^{7}\text{Be}$  by Accelerator Mass Spectrometry

Journal: Atmospheric Chemistry and Physics

**We would like to thank reviewer #1 for the interest shown in our paper and the valuable feedbacks that will improve the quality of the paper. The responses to the comments of the reviewer one by one are as follows:**

**Reviewer #1:** Review of “Detection of stratospheric air intrusion events from ground-based high-resolution  $^{10}\text{Be}/^{7}\text{Be}$  by accelerator mass spectrometry” by X K Liu et al. In this paper, the authors derive  $^{10}\text{Be}$  and  $^{7}\text{Be}$  measurements from collected aerosol and rainwater samples from 7 locations in central China. A large amount of data is presented. I will return to the question of the data coverage in my comments. The results are interesting and should be eventually published in final form, however, some details seems to be glossed over and need to be addressed. In my view, the paper needs major revision.

**Q1:** Abstract, line 30-32. Here, it says that 24% of  $^{10}\text{Be}$  in Xi'an (XA) air was dust-borne, yet the various figures 2 and 3 show considerable variability.

**A1:** We thank the reviewer for the question. Indeed, as the reviewer sees, the dust interference is fluctuating, and the ~24% here refers to the average value. For clarity, it has been modified as follows:

We show that for the city of Xi'an, the average proportion of dust-borne (resuspended)  $^{10}\text{Be}$  was about 24% in 2020/21.

**Q2:** line 48 delete “hard-to-capture”. I am not sure what is meant here, but I assume you mean that it is difficult to quantify the intrusion events. Perhaps rewrite the sentence.

**A2:** We thank the reviewer for the suggestion. "hard-to-capture" has been removed.

**Q3:** line 57 “intrusions”

**A3:** We thank the reviewer for the question, it has been modified:

Traditional observation methods of stratospheric air intrusions mainly rely on....

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**Q4:** line 64 replace “it is generally believed..”with “it has been suggested..”

**A4:** We thank the reviewer for the question, it has been modified:

..., and it has been suggested that ground-based  $^7\text{Be}$  analyses offer a means of identifying and tracing stratospheric air intrusions.

**Q5:** line 74 delete “the” before “very different”

**A5:** We thank the reviewer for the question, it has been modified.

**Q6:** Line 82-87. Here, it is stated that  $^7\text{Be}$  was done by AMS. This is not correct. **Some of the measurements were done by AMS.** In the supplementary data and lines 166-169, it is stated that “some .. membranes were selected and  $\frac{1}{4}$  of them were measured by AMS. The  $^7\text{Be}$  in the remaining  $\frac{3}{4}$  filter membranes...were analyzed by high-purity germanium (HpGe) detector(s)..and analyzed a 477.6KeV..”. Maybe this was only a fraction of the samples, but please rewrite and explain what was actually done.

**A6:** We thank the reviewer for the question. The gamma spectrometry measurement is used here only to correct and check the  $^7\text{Be}$ -AMS measurement results, not to obtain the  $^7\text{Be}$  observation record. All the observational records of  $^7\text{Be}$  and  $^{10}\text{Be}$  presented in this work are obtained based on AMS measurements. For clarity, the following description has been added on the first sentence of the last paragraph of section 2.3:

In addition to  $^7\text{Be}$  being measured by AMS, we also correct and cross-checked the results of  $^7\text{Be}$ -AMS by gamma spectroscopy.

**Q7:** Line 105-106. It is stated here that “an annual record with daily resolution” was obtained. This is most true for site XA (Xi'an) and QL (Qinling) but even there, it's not actually daily, there are missing values.

**A7:** We thank the reviewer for the comment. The daily resolution here means that the sample sampling period is one day, some of which are continuous every day, and another part of the time is not continuous due to the sampling process such as impact of COVID-19.

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**Q8:** Section 2.1 Samples from the other sites were only measured at a few times in October 2020, Jan 2021 and April 2021.

**A8:** We thank the reviewer for the question. The resuspension of terrestrial dust is controlled by aridity and wind power. Therefore, we chose representative sampling sites with different atmospheric relative humidity and NDVI characteristic to verify the general applicability of this calibration method. The distribution of sample collection times at these sampling sites is noted at the appropriate place in the manuscript:

Zhongwei (ZW, 37°30'57"N, 105°11'8"E), Taiyuan (TY, 37°48'26"N, 112°34'55"E), Chengdu (CD, 30°56'44"N, 103°40'41"E), Nanning (NN, 22°50'18"N, 108°16'51"E), and Lianyungang (LYG, 34°46'4"N, 119°26'55"E) for short-term intermittent monitoring (in October 2020, Jan 2021 and April 2021).

**Q9:** Section 2.3 lines 148-151 and supplement. 1-2. Here, it is stated that the "transmission after the second stripping foil" was improved to ~24%. If this is into  $^{10}\text{Be}^{4+}$ , this is a remarkable improvement. Most labs get lower yields even into  $3+$ . Also, what is the location of the second stripping foil? I assume it is after the analyzing magnet and before an ESA, but no details of the AMS setup are given. The discussion in the reference Zhang and Fu (2017) is not completely helpful. First, in that paper, the yield from  $\text{Be}^{2+}$  to  $\text{Be}^{3+}$  is given as 31% **and from  $\text{Be}^{3+}$  to  $\text{Be}^{4+}$  is 3%. If some other improvement was done, it is not clear from this paper.** Also, much of the paper of Zhang and Fu (2017) discusses other methods, such as using  $\text{BeF}_3^-$  as an injection beam. Hence, we still don't know from the paper presented here by Liu et al. which method was used.

**A9:** We thank the reviewer for the question. In this work, the  $\text{BeO}^-$  to  $\text{Be}^{2+}$  to  $\text{Be}^{4+}$  method is used to perform  $^{10}\text{Be}$ - $^7\text{Be}$ -AMS measurements (Zhang and Fu, 2017). Secondary stripping film (500 nm  $\text{Si}_3\text{N}_4$ ) behind the  $115^\circ$  high energy analysis magnet. In most instances, the efficiency of  $\text{Be}^{2+}$  to  $\text{Be}^{3+}$  is ~31% ( $^{10}\text{Be}$  standard sample) (Zhang and Fu, 2017).

The low efficiency of  $\text{Be}^{2+}$  to  $\text{Be}^{4+}$  is due to the establishment of our preliminary method in 2017, and the components behind the stripping film have not been carefully optimization. In particular, the role of the quadrupole lens behind the  $65^\circ$  ESA is underestimated. This work was carried out after the  $^7\text{Be}$ -AMS transmission efficiency

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was re-optimized in 2018. That is to say, the most important thing is to adjust that quadrupole lens (although the transmission efficiency is lower in the early work of 2017, AMS is a relative measurement, which ultimately only affects the statistical error of the final results). Based on this, in the  $^{10}\text{Be}$  analysis, the terminal voltage works at 2.5 MV, the efficiency of  $^{10}\text{Be}^{2+}$  to  $^{10}\text{Be}^{3+}$  is  $\sim 31\%$ , and the efficiency of  $^{10}\text{Be}^{2+}$  to  $^{10}\text{Be}^{4+}$  is  $\sim 24\%$ . An example from the same standard samples in this work, AMS measurements of  $^{10}\text{Be}^{3+}$  and  $^{10}\text{Be}^{4+}$  in the  $^{10}\text{Be}$  standard sample ( $^{10}\text{Be}/^9\text{Be} = 2.709 \cdot 10^{-11}$ ) are given below:

Table 1 AMS measurement raw results of  $^{10}\text{Be}^{3+}$  and  $^{10}\text{Be}^{4+}$  in  $^{10}\text{Be}$  standard samples of this work ( $^{10}\text{Be}/^9\text{Be} = 2.709 \times 10^{-11}$ )

No.	Measurement No.	Raw average of $^{10}\text{Be}^{3+}$	Efficiency <sub>1</sub>	Raw average of $^{10}\text{Be}^{4+}$	Efficiency <sub>2</sub>
1	Be2020-0313	$8.51 \times 10^{-12} \pm 4.25 \times 10^{-14}$	31.41%	$6.46 \times 10^{-12} \pm 3.20 \times 10^{-14}$	23.85%
	Be2020-0314	$8.51 \times 10^{-12} \pm 5.20 \times 10^{-14}$	31.41%	$6.54 \times 10^{-12} \pm 3.48 \times 10^{-14}$	24.14%
2	Be2021-737	$8.53 \times 10^{-12} \pm 6.63 \times 10^{-14}$	31.78%	$6.70 \times 10^{-12} \pm 3.91 \times 10^{-14}$	24.73%
	Be2021-738	$8.50 \times 10^{-12} \pm 5.98 \times 10^{-14}$	30.79%	$6.74 \times 10^{-12} \pm 3.64 \times 10^{-14}$	24.88%
		<b>Mean</b>	<b>31.35%</b>	<b>Mean</b>	<b>24.40%</b>

Corresponding instructions are added in Supplementary Information:

### Analysis of $^7\text{Be}$ and $^{10}\text{Be}$ by AMS

First, both were extracted as  $\text{BeO}^-$  particles from the ion source. Next, with the accelerator terminal voltage set at 2.5 MV, beryllium ions are accelerated and the +2 charge state is selected using a high-energy analysis magnet. Stripping efficiency of  $\text{BeO}^- \rightarrow \text{Be}^{2+}$  is approximately 47%. After the stable and rare nuclides were separated by the analysis magnet, the rare nuclides passed through a secondary stripping film (500 nm,  $\text{Si}_3\text{N}_4$ ) are stripped to +4 charge state. The beryllium atoms are completely stripped, that is,  $\text{BeO}^- \rightarrow \text{Be}^{2+} \rightarrow \text{Be}^{4+}$ . Based on the  $\text{Be}^{2+}$  to  $\text{Be}^{4+}$  method we initially established in 2017 (Zhang and Fu, 2017), the stripping efficiency has been improved. The increase in efficiency is based on the carefully adjustments to the quadrupole lens behind the  $65^\circ$  ESA. Based on this, in the  $^{10}\text{Be}$  analysis, the terminal voltage works at 2.5 MV, the efficiency of  $^{10}\text{Be}^{2+}$  to  $^{10}\text{Be}^{3+}$  is  $\sim 31\%$ , and the efficiency of  $^{10}\text{Be}^{2+}$  to  $^{10}\text{Be}^{4+}$  is  $\sim 24\%$ .....

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**Q10:** Line 182-188. Here, the values from previous work on  $^{10}\text{Be}$  have errors, it would be good to quote them here.

**A10:** We thank the reviewer for the suggestion. Here, we choose the mean value within the range of reported values in papers in this area as the topsoil  $^{10}\text{Be}$  concentration and cite their results. In addition, the choice of the topsoil  $^{10}\text{Be}$  concentration value had little effect on the correction results, which mainly depends on the resuspension degree of soil dust (this is mainly calculated by the content of Al in the aerosol).

**Q11:** Line 234. "material"

**A11:** We thank the reviewer for the suggestion, it has been modified:

...and northern deserts profoundly impact the supply of aeolian material on the Loess Plateau.

**Q12:** Line 236. I think "atmospheric movement information" isn't a good term, please use something like "atmospheric circulation"

**A12:** We thank the reviewer for the suggestion. Modify it to atmospheric circulation: obscures degree of  $^{10}\text{Be}$  change brought by atmospheric circulation such as STE (Fig. 1a-c).

**Q13:** Line 247. I think the accepted value of the half-life is 1.38Myr.

**A13:** We thank the reviewer for the suggestion, its half-life has been revised to 1.39 (1.38) Myr:

Due to its long half-life ( $1.39 \times 10^6$  years).....

**Q14:** Line 250. "cannot"

**A14:** We thank the reviewer for the suggestion, it has been modified:

$^7\text{Be}$  cannot accumulate in the soil due to its relatively short half-life (53.3 days)

**Q15:** Line 265 "aridity" not "dryness"

**A15:** We thank the reviewer for the suggestion, it has been modified:

.....by aridity and wind power.

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**Q16:** Figures 2,3 and 4 have small font sizes and they can be hard to read. I suggest scaling up the figures in the revised version.

**A16:** We thank the reviewer for the suggestion. The font sizes for Figures 2, 3 and 4 have been enlarged.

**Q17:** Line 333-334. What are the "V" events and don't you have meteorological data about what happened on those days at XA and QL?

**A17:** We thank the reviewer for the question. The "V" event mentioned here refers to the change in the observed aerosol  $^{10}\text{Be}/^{7}\text{Be}$  ratio in the days before, during and after the rain (enlarged view of Fig. 3a). That is, the  $^{10}\text{Be}/^{7}\text{Be}$  ratio was high in the days before the rain, it decreased significantly during the rain, and the value began to rise again in the days after the rain stopped. This also shows that the interference of resuspended dust is relatively large in the loess area of China, and correction is necessary. We have supplemented this as follows:

This fluctuation is especially apparent at the XA site and is seen as "V"-shaped changes in the uncorrected record (enlarged view of Fig. 3a, which refers to the uncorrected aerosol  $^{10}\text{Be}/^{7}\text{Be}$  ratios change in the days before, during, and after the rain).

**Q18:** Line 402-406 restates what has already been said and can be deleted.

**A18:** Thanks to the reviewer for the suggestion, which has been removed.

**Q19:** Line 369. This seems to be a new subject here. It appears the authors try to argue that the ozone in the ground-based measurements at XA are not due to photochemical effects (e.g. due to pollution) and the ozone is due to stratospheric injection. This seems like a long shot, especially in a city like XA with considerable anthropogenic air quality problems, although I agree others (e.g. Langford) have assert similar effects elsewhere.

**A19:** We thank the reviewer for the question. We do not deny the contribution of photochemical effects to surface ozone here, but want to express that the contribution of stratospheric ozone input in this period is more obvious than that in other seasons. In this work, for the seasonal intrusion process confirmed by other means, we reveal that the intrusion of stratospheric ozone in spring is significantly higher than in other seasons by the phenomenon that the  $^{10}\text{Be}/^{7}\text{Be}$  ratio in spring is significantly higher

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than that in other seasons (~3 times). We are sorry that our statement misled the reviewer's understanding. To give a clearer expression, modify the description in this paragraph as follows:

Sunlight is a natural prerequisite for the photochemical generation of O<sub>3</sub> at the earth's surface (Kondratyev and Varotsos, 1996). The sunlight radiation follows a symmetrical intensity law in the spring and autumn at the XA site (Fig. 4c). However, surface O<sub>3</sub> concentrations at the XA site (Fig. 4b) are inconsistent with regularity of sunlight radiation intensity (mainly in the spring represented in Fig. 4b blue area). This feature is consistent with the seasonal increase in <sup>10</sup>Be/<sup>7</sup>Be. We suggest that the abnormally increased part of the surface O<sub>3</sub> concentration may largely reflect the influx of stratospheric O<sub>3</sub>, indicated by frequent and relatively large changes in the <sup>10</sup>Be/<sup>7</sup>Be. Similar seasonal intrusion results have been confirmed during spring in other regions using ground-based lidar observations (Langford et al., 2009), airborne observations (Weigel et al., 2012), and atmospheric models (Zhao et al., 2021).

**Q20:** Figure 4b. How are the data in figure 4b compared to fig. 4a? Are all the data used? 4b refers only to XA, but 4a also includes QL data.

**A20:** We thank the reviewer for the comment. Figure 4b refers to XA data only. The straight-line distance between XA and QL is about 60 km (described earlier in Section 3.1). The focus here is on stratospheric mass intrusion processes with large scales. In addition, there is no significant difference in the seasonal variation scale of surface ozone concentration and sunlight intensity between the two site. Therefore, only a weather station was set up in XA to collect solar radiation intensity data, and the ozone data at XA was downloaded from the China Meteorological Network. Added clarification in the legend to Figure 4 as follows:

**Fig. 4** .....Note: XA and QL are ~ 60 km apart, so the seasonal changes of sunlight radiation and ozone are represented by the XA site data.

**Q21:** Line 455. "STE is an important channel...". This sentence needs rewriting and makes little sense at present. Perhaps STE are important processes that can quickly transmit chemical material from the stratosphere into the lower atmospheric environment.."

**A21:** Thanks to the reviewer for the comment and suggestion. Modify the content as

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suggested:

STE are important processes that can quickly transmit chemical material from the stratosphere into the lower atmospheric environment, particularly low-intensity and frequent stratospheric air intrusion events that can transmit chemical matters more quickly to affect the environment.