Interactive comment on “Airborne radionuclides and heavy metals in High Arctic terrestrial environment as the indicators of sources and transfers of contamination” by Edyta Łokas et al.

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We are thankful for the thorough and constructive comments and remarks on our manuscript. The issues raised by the reviewer were taken into consideration and in the following paragraphs, we present our reply to each of them. Below we provide our responses.

Anonymous Referee #2: General comments: From an analytical perspective, this article represents a great amount of work with regard to sample collection and processing, chemical and radiochemical separations, instrumental measurements, and interpretation of results. A systematic organization of the measurement results is found in the extensive tables of analytical data; these will enable valuable comparisons with past and future data obtained by other researchers in that region. Perhaps referring to the tables, figures and plots with very brief descriptions should comprise the “Results” section. I’m not sure that it is necessary to mention the ranges of all the analytes in the text when this information can be seen in the tables. We agree with the reviewer that the results section should be modified. The results for soils and cryoconite were linked. The results section was divided for two subsections: “Radionuclide contents and their activity ratios in tundra soil profiles and in cryoconite samples” and “Heavy metals analyses in tundra soil profiles and in cryoconite samples”. Generally, this section was shortened, figures were merged and the author selected airborne radionuclides in soils and cryoconites and only two heavy metals in soils and cryoconite and presented on Fig 2 and 3, respectively. We will present additional data on radionuclides in cryoconites (some of that are not published yet) in the corrected Fig 5 (now Fig 6). In the results section we left only maximum value for radionuclides and only average values for isotopic ratios. For heavy metals we left ranges because Figure 3 presents only concentrations of two metals: Pb and Cd. We removed other heavy metals from the graph because these data are presented in supplementary material.

The “Discussion” section is partitioned into three subsections. Subsection 4.2 “The source of radionuclides and heavy metals contamination” is essentially entirely devoted to radionuclides; perhaps it would be better just to drop the “heavy metals” part from this title since it is discussed in depth in subsection 4.3. Some of the actual discussion of the results, especially for stable heavy metals and correlations among them (subsections 3.3 and 3.4), occurs in the “Results” section. It is recommended to integrate these remarks into the “Discussion” section along with the existing material. The discussion was modified, it was merged and divided only for 2 subsections: “4.1. Radionuclides
and heavy metals contamination" and “4.2. The source of radionuclides and heavy metals contamination”. Most of the raw data of radionuclides and heavy metals in the discussion were removed.

Uncertainties are presented with the tables S2 through S5 giving radionuclide measurement results; however, I cannot find any mention in the text of whether these uncertainties are at the 68% (k=1) or 95% (k=2) confidence level. A general statement about the confidence level of uncertainties should be given early in the text (e.g., at the very beginning of the “Results” section). In addition, the main contributions to the radioanalytical uncertainties should be mentioned (e.g., counting statistics, calibrations, tracer concentrations, etc.). The information about uncertainties and confidence level was added to the Methods section: “The uncertainties of activity concentrations include the measurement uncertainties (counting statistics and efficiency) and are reports as 1 \( \sigma \) counting statistics.”

Table S6 lists all of the stable element measurement results, but there are no uncertainties attached to these values. A statement of uncertainty “ranges” associated with each element concentration measurement should be given for the reader, perhaps at the column headings or as a footnote. The information about uncertainties was added to the “Methods” section: “The quality control and assurance for this method is presented in Zaborska et al. (2017).” My recommendation is that this article should be published with minor revisions as outlined above.

Specific comments to revised article: 1. Although I believe Svalbard and Spitsbergen refer to the same archipelago, the authors should choose one or the other to be used consistently in the article. The authors choose Spitsbergen.
2. In the abstract, it would be useful to mention that Kaffiøyra region is located in Spitsbergen “Spitsbergen” was added.
3. In subsection 2.3.2 “Heavy metal analyses” it is not mentioned if separate subsamples (from those used for radionuclide analyses) were used for stable element determinations. If separate samples were used, how much material and what dissolution methods were used to prepare them for the flame atomic absorption and mass spectrometer measurements? Following text was added: “Separate sub-samples (0.5 g) were dedicated for measurements of selected heavy metals that are believed to be important anthropogenic contaminants (Pb, Cu, Zn, Cd). Contaminating metals and other metals (Fe, Al, Ni, Cr, Mn) were measured with a flame atomic absorption spectrometer (AAS Shimadzu 6800) using deuterium background correction.”
4. How was sample organic content assessed and related to LOI (which needs to be defined in the text)? It was defined in the “Methods” section: “Organic matter contents were determined by loss on ignition (LOI) at 600° C for 6 hours.”
5. The plots of depth distribution in fig 2 are important but very difficult to read; a significant improvement in quality is needed. Figures 2 and 3 were merged and the authors selected airborne radionuclides and selected heavy metals and presented on new Fig 2 and 3.
6. The reference “Łokas et al., 2010” in line 10 on page 5 is missing. It was added to the “References” section.
7. In line 6 on page 17, “average mass ratios” should be “average atom ratios” to be consistent with all of the previous text. Corrected.
8. In line 8 on page 17, “…initial soils from other west cost of Spitsbergen” is confusing. It was corrected as “…initial soils from other west coast of Spitsbergen”.
9. In line 8-9 on page 17, perhaps to write “We observed an excess of 238Pu in cryoconite samples similar to that which we observed in other initial soils in Spitsbergen, and this enrichment of 238Pu is derived from sources other than 239Pu. This is due to pure 238Pu from a satellite re-entry after injection from the stratosphere into the troposphere.” Corrected.
Fig. 1. Location of the study area. Profile S01-S06 were collected at increasing distances from the front of Waldemarbreen and cryoconite samples (2-13) from different altitudes of this glacier.
Fig. 2. The model of Waldemarbreen with the activity concentration of atmospheric radionuclides (210Pb, 137Cs, 239,240Pu, 241Am) in cryoconite and depth distribution of these radionuclides in tundra soils.

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Fig. 3. Concentrations of selected heavy metals (Pb, Cd) in cryoconite samples and tundra soil profiles.

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**Fig. 4.** The contribution of global fallout Pu calculated using plutonium isotope ratios (left) contribution of anthropogenic origin Pb calculated using Pb isotopes and end-member method.

**Fig. 5.** The relation between the 240Pu/239Pu atom ratios and the 238Pu/239+240Pu activity ratios in investigated cryoconite samples, other Arctic cryoconite (Łokas et al., 2018 and unpublished data) and cryoc