Response to Interactive comment from Anonymous Referee #1

Interactive comment on "Temporal variation of ^{129}I and ^{127}I in aerosols from Xi'an, China: influence of East Asian monsoon and heavy haze events" *by* Luyuan Zhang et al.

Anonymous Referee #1

Received and published: 19 October 2019

The paper acp-2019-818-manuscript-version1 entitled "Temporal variation of 129I and 127I in aerosols from Xi'an, China: influence of East Asian monsoon and heavy haze events" provides interesting data for the distribution of iodine isotopes in the aerosols of part of China which are missing from the international data base. The paper has the potential for publication after revision as given below.

We are very grateful for the reviewer's positive and constructive suggestions and comments that make our manuscript better. According to the reviewer's comments, the main modifications are made in the revised version:

1) The language is further polished, including revising the grammar mistakes and shortening the long sentences;
2) The Figs 1, 3, S1, S2, S7 and S8 are reorganized and adjusted to make the discussion part more easily understood and convincing.

The comments are responded item by item as below.

1. The paper needs some linguistic revision as there are many grammatic mistakes.

Response: Sorry for such basic mistakes. We have checked throughout the context and revised all the linguistic mistakes.

2. Lines 44-45 "As a consequence of these point sources of 129I, the distribution of 129I is rather uneven (Snyder et al., 2010)? Where?

Response: These point sources of ^{129}I have been listed in the reference of Snyder et al., 2010, including the principal nuclear reprocessing plants in Russia, UK, France, USA, Pakistan, China, Israel, India, South Africa and Argentina, nuclear accidents in Chernobyl, Former Soviet and Fukushima, Japan. For brief introduction, only references are cited here, not listing all these specific sources. This sentence has been revised to be "As a consequence of ^{129}I releases from NFRPs, nuclear accidents and nuclear weapon testing sites, the global distribution of ^{129}I is rather uneven (Snyder et al., 2010; Xu et al., 2015)."

3. The paragraph between lines 50 and 55 is long and difficult to follow and could be rewritten to focus on aims of the study.

Response: Line 50 has been separated into two parts, and revised to be "And those previous studies present the time series of ^{129}I in aerosols in monthly resolution for the purpose of

nuclear environmental monitoring. Such a low time-resolution is not sufficient to understand the source, transport and temporal variation pattern and its influencing factor of ^{129}I."

Line 51-55 paragraph has been reorganized into three sentences as below.

"Here, we present a day-resolution temporal variation of ^{129}I and ^{127}I in aerosols during 2017/2018 from a typical monsoonal zone, Xi'an city in the Guanzhong Basin of northwest of China, to make attempts to investigate the level, sources and temporal change characteristics of ^{127}I and ^{129}I. This study will help to establish a background value of $^{129}I/^{127}I$ ratio serving the nuclear environmental safety monitoring. The possible influencing factors on temporal variation of iodine isotopes are also explored, including meteorological parameters, East Asian monsoon (EAM) and heavy haze events."

4. It is not clear how much time is the "a day-resolution" sampling reflects in term of iodine residence time in the atmosphere?

Response: Iodine residence time is closely related to its species and the associated states with particles, generally ranging from a few seconds to a few days for gaseous iodine species (Saiz-Lopez et al., 2012). For particle-associated iodine, its residence time is much dependent on particle size, varying from 0.1 day to 10 days (< 1 day for particles large than ~ 1 μm, and > 1 day for particles smaller than ~ 1 μm) (Moyers and Duce, 1972). In this study, total suspended particles were sampled in a 24 h resolution, reflecting a relatively equilibrated state for gaseous-particle converted iodine and large particle-associated iodine with shorter residence time (< 1 day), and a more variable information of iodine in small particles. Such a time-resolution can benefit greatly our future work on speciation analysis of atmospheric iodine to dive deep into atmosphere iodine processes.

Referece:

Moyers, J.L., Duce, R.A., 1972. Gaseous and particulate bromine in the marine atmosphere. J. Geophys. Res. 77, 5330–5338. https://doi.org/10.1029/jc077i027p05330

Saiz-Lopez, A., Gómez Martín, J.C., Plane, J.M.C., Saunders, R.W., Baker, A.R., Von Glasow, R., Carpenter, L.J., McFiggans, G., 2012. Atmospheric chemistry of iodine. Chem. Rev. 112, 1773–1804.

5. Figures 1 a and b can be combined in one figure.

Response: Figure 1 has been reorganized. Figure 1b as an inset has been combined with 1a.

6. The results part needs further additions from the supplementary data including Figs. S1 and S2.

Response: The results including Figs. S1 and S2 in the supplementary data have been added into the manuscript.

7. Connection of iodine chemical forms (I-127 and I-129) from the sources and in the atmosphere may elucidate some of the inconclusive correlations and relationship to spatial and temporal atmospheric transport on short and long distances.

Response: We strongly agree with this comment. The short- and long-range transport of airborne iodine is strongly related with its chemical forms. The present study focuses on aerosol iodine, and the gaseous iodine (^{127}I and ^{129}I) sample collection and analysis are under way, while no data has been available at present. We do expect that further work on iodine chemical forms in air would give further understanding on the relationship.

8. More elaboration of weathering of basement rocks as a source of I-129 will be interesting.

Response: In line 226-236, more explanation has been added as below. "Weathering of bed rock is not also a major source of airborne ^{129}I, since weathering just contributes 5% of stable iodine, and ^{129}I in bed rock can be considered even lower the nature-produced ^{129}I level because of the continuous decay."

9. Addition of Figure S7 and S8 to the discussion section will enhance the understanding of the atmospheric transport pathways of the isotopes.

Response: For improving the understanding, Figure S8 and SI-4 discussion part have been moved to the manuscript. Considering too many figures and each importance, Figure S7 is still kept in the Supplementary Information.

10. More details on the paragraph in lines 100-104 can add clarity to general statement with respect to 127I distribution in China.

Response: Line 100-104 have been added more discussion of aerosol iodine in China. "Whereas, a similar range of TSP ^{127}I was observed to be 4.5-22 ng m^{-3} at coastal urban, Shanghai, China, and iodine concentration were lowest in summer and an increase occurred in fall and winter (Gao et al., 2010). Iodine associated with PM10 and PM2.5 were found to be 3.0-115 ng m^{-3} and 4-18 ng m^{-3}, respectively, in urban and island sites of Shanghai, slightly lower than TSP iodine (Cheng et al., 2017; Gao et al., 2010). The marine aerosol iodine offshore China was found below 8.6 ng m^{-3} during the XueLong cruise from July to September 2008 (Xu et al., 2010). These results suggest a relatively high aerosol ^{127}I level in both inland and coastal urbans in China."

Referece:

Cheng, N., Duan, L., Xiu, G., Zhao, M., Qian, G., 2017. Comparison of atmospheric PM2.5-bounded mercury species and their correlation with bromine and iodine at coastal urban and island sites in the eastern China. Atmos. Res. 183, 17–25. https://doi.org/10.1016/j.atmosres.2016.08.009

Gao, Y., Sun, M., Wu, X., Liu, Y., Guo, Y., Wu, J., 2010. Concentration characteristics of bromine and iodine in aerosols in Shanghai, China. Atmos. Environ. 44, 4298–4302. https://doi.org/10.1016/j.atmosenv.2010.05.047

Xu, S., Xie, Z., Li, B., Liu, W., Sun, L., Kang, H., Yang, H., Zhang, P., 2010. Iodine speciation in marine aerosols along a 15000-km round-trip cruise path from Shanghai, China, to the Arctic Ocean. Environ. Chem. 7, 406–412.

11. The anthropogenic source for I-127 is mainly related to coal consumption (local source) whereas the I-129 source is mainly related to far away transport. It will be good to provide some details of how these isotopes are associated in the atmosphere with respect to airmasses altitude, chemistry and residence time of the isotope.

Response: The mechanism for iodine association with particles with many uncertainties, generally has two pathways, iodine compounds as primary nuclei during fine particle formation, and adsorption onto naturally occurring particles (Garland, 1967; Saiz-Lopez et al., 2012). In section 4.2.2, association of ^{127}I and ^{129}I with particles has been elucidated in respect of nucleation process and residence time. However, some interpretations need more evidence. Chemical processes could definitely affect iodine species in aerosols, such as inorganic iodide and iodate, the former of which has been found in a large proportion of inorganic iodine likely because of the presence of reductant SO_3 (Zhang et al., 2016). As this paper focuses on the temporal variation of total iodine isotopes and there is no more data to suggest that chemical process would significantly affect the total iodine change, therefore, it is hard to discuss the influence of chemical processes on iodine bound to particles. Our future work on aerosol iodine species will put more efforts on this point. In section 4.2.3, "Furthermore, the back trajectory analysis also showed that the low ^{129}I level on April 18 can be partially attributed to an ^{129}I-poor low-altitude air mass (< 900m) (Fig.S3a), since either they might be formed in ^{129}I-poor inland areas, not from the ^{129}I-rich European area, or long-range transported ^{129}I in low-altitude air mass could be easily lost by the topographic countercheck (Dong et al., 2018)." has been modified to explain the influence of airmass altitude.

Referece:

Dong, Z., Shao, Y., Qin, D., Zhang, L., Hou, X., Wei, T., Kang, S., Qin, X., 2018. Insight Into Radio-Isotope 129I Deposition in Fresh Snow at a Remote Glacier Basin of Northeast Tibetan Plateau, China. Geophys. Res. Lett. 0. https://doi.org/10.1029/2018GL078480

Garland, J.A., 1967. The adsorption of iodine by atmospheric particles. J. Nucl. Energy 21, 687–700.

Zhang, L., Hou, X., Xu, S., 2016. Speciation of 127I and 129I in atmospheric aerosols at Risø, Denmark: Insight into sources of iodine isotopes and their species transformations. Atmos. Chem. Phys. 16, 1971–1985.

12. May be good to make the text in Figure 3 in larger font.

Response: The font in Figure 3 has been adjusted larger.

13. Although the authors pointed out the possible use of air masses transport to predict iodine sources and impact on future iodine distribution, it is still not clear how the iodine data enhance our understanding of the climate or atmospheric circulation.

Response: Here we use a set of day-resolution iodine isotopes data to establish the crucial linkage with East Asia monsoon system and meteorological conditions. On this basis, long-term observation of iodine isotopes would refresh the understanding of climate change, not just one-year short-term iodine data presented in this paper. Moreover, high time-resolution

iodine data in combination with modelling should be expected. Before make the application on climate and atmospheric circulation, plenty of questions have to be well answered, for instance, how airborne iodine species like and interact with each other.