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Overall I think Sierra-Hernandez and others present a comprehensive and interesting follow-up of the 1992 Guliya ice core and highlight some important trends in trace metals that have taken place since then. In general I only have minor comments and suggestions. The largest remaining question in my mind is how an NAO index is mechanistically related to higher EFs, so an explanation of that would be helpful.

Line 56- What is the “new Silk Road”?

[Response] It is an initiative proposed by President Xi Jinping formally called the Belt and Road Initiative to modernize and build new railways, ports, pipelines, power grids and highways to connect China with the west. This has been added accordingly.

Line 65- Is this percentage (50-60%) meant to imply this is how much emissions can be attributed to motor vehicles?

[Response] Yes, we have changed it to “(estimated to be ~60–70 % of the total air emissions)”. There was also a typo and it should be 60-70 % and not 50-60 %.

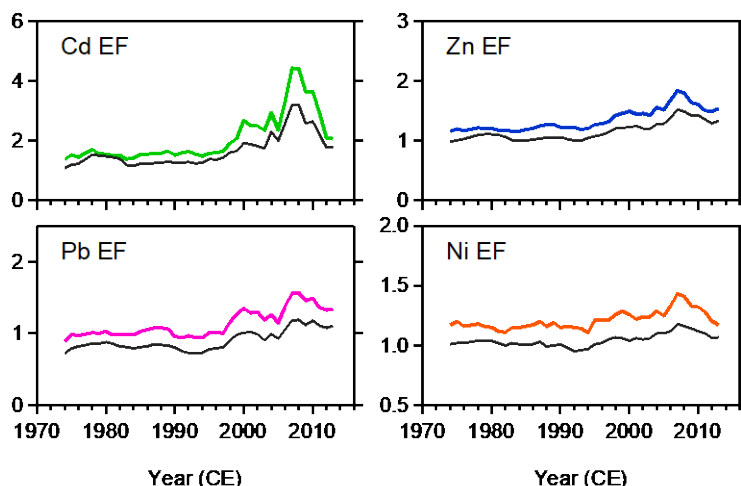
Lines 93-95- If more anthropogenic sources have emerged, then what new developments have taken place and are in use in this manuscript to attribute possible sources?

[Response] New comprehensive emission inventories of air pollutants have been developed since the 1970s and 1980s to regulate their emissions. In particular, we use the Emissions Database for Global Atmospheric Research (EDGAR v4.3.2) which compiled a comprehensive dataset of air pollutants between 1970 and 2012 and is used in this manuscript to attribute possible sources along with emission inventories from the U.S. Energy Information Administration, and the British Petroleum (BP) Statistical Review of World Energy. In our previous publication (Sierra-Hernandez, 2018), the TE ice core record stopped at 1991, and thus the increases observed between 1975 and 1991 were not possible to compare with emission inventories since the time interval was too short. With this new Guliya core drilled in 2015, we now have a 40-year timeseries of TEs that can better show temporal trend similarities with emission inventories. A description of these new developments is now included in the text.

Lines 139-140- Fe is an interesting choice. Why not something else like Al? Or Ti?

[Response] In our previous publication (Sierra-Hernandez, 2018) we used Fe as a crustal element. To be consistent, we continue to use Fe as a crustal element in this new ice core record. In lines 143-145 we explain that “Fe was chosen as the crustal reference due to its stability and high abundance in soil and rocks (Wedepohl, 1995), its high concentration both in the ice core samples and the PSAs, and the ability of the ICP-SFMS to measure Fe with high accuracy and precision”. We added the following text in lines 149-152: “Additionally, Fe is highly correlated with Al ($r = 1$), and also with Ba ($r = 0.98$). Like in Sierra-Hernandez (Sierra-Hernández et al., 2018), EFs calculated using Al and Ba as crustal references had no significant differences compared to EFs relative to Fe which all together shows that the choice of Fe as a crustal TE to calculate EFs did not affect the results.”

Here is the comparison between EF relative to Fe (color curves) with EFs relative to Al (black curves)



Lines 150-152- What is defined as “pre-industrial” in this case? Before what time period?

[Response] We consider the pre-industrial period to be the period before the Industrial Revolution in Europe (~1780). Similar to our previous publication (Sierra-Hernandez, 2018), we used the period 1650-1750 as pre-industrial to be more conservative due to the time scale uncertainty of the ice core. We have clarified this in the text accordingly.

Lines 192-194- I understand the difference between EF and Excess calculations, but why would some elements have significant trends for EF but not Excess? This needs some additional explanation.

[Response] For all our statistical tests we used the datasets at full resolution. However, the Mann-Kendall trend tests were performed with the annual dataset. The problem with the Excess concentration was that by averaging the values within one year, large negative values skewed the final results to negative numbers. The tests have been re-done using the full resolution EF and Excess concentration datasets. The updated results show that both EFs and Excess concentrations of Bi, Cd, Ni, Pb, Tl, and Zn show significant increasing trends. Changes were made in the manuscript accordingly.

Lines 224-225- Since the EDGAR database excludes biomass burning and land use change, is it possible that some of the observed trends could be attributed to these processes? I know it will be hard to make these estimations without quantitative data, but these could be potentially significant.

[Response] The EDGAR database does include some anthropogenic biomass burning activities such as domestic combustion and agricultural waste burning excluding only large-scale biomass burning. We have added a Biomass Burning section (Sect. 3.1.2) to discuss domestic combustion and agricultural waste burning.

It is possible that the TE trends observed in the Guliya ice core can be attributed to large-scale biomass burning and land use change. Recent increases in wildfires in the Himalayas have been detected in Central Tibet possibly due to recent warming (You et al., 2018). This is mentioned in Sect. 3.1.2.

Lines 303-305- A brief mechanistic explanation about how a positive NAO index actually results in higher EFs would be useful.

[Response] We have eliminated Section 3.2. Please see the full response regarding this in the responses to Reviewer #2.

Lines 310-313- However, I think it’s important to note that the drop from 1940-1970 is also when there is a gap/transition in the data. Neither coal nor oil consumption estimates extend fully back

through this period. And while coal production is still reasonably high, it does start to decline. This is not to say that I don't believe the NAO is having an impact, but I think it's important to highlight that there are some gaps in the data.

[Response] Yes, we agree that the gaps in the inventories are important and should be noted. As described above the NAO discussion has been eliminated, therefore these inventories are not shown/discussed anymore.

Lines 337-336- This is an interesting development and would seem to be a reversal of coal consumption declines since 2009 as previously noted in line 320. Was the decline in 2009 a temporary slow down then?

[Response] Yes, it is a temporary slow-down between 2009 and 2013. This Figure has been eliminated.

Figure 5- The second y-axis on the right needs a label.

[Response] Label has been added. The Figure was moved to the Supplement as Fig. S6

Figure 6- Why is the 1992 core plotted as a 5-year running median while the 2015 core is a 5-year running mean?

[Response] The 2015 Guliya EF composite z-scores were plotted as 5-year running means in Fig. 6 to keep consistency with the previous figures of the manuscript. Since Section 3.2 has been removed, there is no Figure 6 anymore.

References

You, C., Yao, T., and Xu, C.: Recent Increases in Wildfires in the Himalayas and Surrounding Regions Detected in Central Tibetan Ice Core Records, J. Geophys. Res.: Atmospheres, 123, 3285-3291, 10.1002/2017jd027929, 2018.