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Title: Comment on: "A novel approach to peatlands as archives of total cumulative spatial pollution loads from atmospheric deposition of airborne elements complementary to EMEP data: priority pollutants (Pb, Cd, Hg)" by Ewa Miszczak, Sebastian Stefaniak, Adam Michczyński, Eiliv Steinnes and Irena Twardowska.

Article Type: Letter to the Editor

Keywords: Lead, immobility, ombrotrophic peatland, bog, geochemistry, enrichment factor, metal accumulation rate

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Subject: Critical Comment on Miszczak et al (2020) – R2

Buenos Aires, April 6th, 2020

To Science of the Total Environment Co-Editors in chief Damià Barceló and Jay Gan.

Dear Dr Barcelo and Dr Gan,

We hereby submit our revised version (R2) of our letter to Editor to comment the paper that Miszczak et al. (2020) recently published in Science of the Total Environment (https://doi.org/10.1016/j.scitotenv.2019.135776).

We reiterate the importance of the mistakes those authors have made, together with ignoring the vast majority of literature in the field. This paper should not have passed the review process and while reviewer 1 claims our tone is insulting and attacking, we really don't think so. If any, it is quite the opposite as our community has felt attacked and insulted, otherwise we would not be 27 to co-sign this comment. As such, this paper is neither a research group argument nor a discussion. It is just a community putting straight back the many major flaws of this paper, for the sake of scientific accuracy and for the STOTEN readers.

We kept out short Supplementary Material that gives the opportunity to list the entire literature as we think it is crucial to show how many papers are contesting what Miszczak et al. (2020) have claimed and that our community finds it imperative to correct their many scientifically invalid claims. As it contains other corrections and criticisms, we do not think it could be appropriately converted in a "data in brief format", but are happy to further discuss that point with you.

We reaffirm that the approach that the peat geochemistry community follows to reconstruct past anthropogenic activities using lead profiles in ombrotrophic peat has been the pillar of most of the papers dealing with Pb as an indicator of global pollution over the world as you can see in the table in the Supplementary Material.

We insist on the fact that the vast majority of our community was appalled by the publication of Miszczak et al. (2020) -as the reviewers point out this paper should not have passed the revision- and have therefore co-signed this Letter to Editor. We find it fundamental to publish it. We therefore hope you are satisfied with this version of our comment that we hope to be final.

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On the behalf of: Baron S. (CNRS, Toulouse, France), Cloy J.M. (Scotland's Rural College, Edinburgh, UK), Enrico M. (Harvard, USA), Ettler V. (Charles University Prague, Czech Republic), Fagel N. (U. Liège, Belgium), Kempter H., Kylander M. (U. Stockholm, Sweden), Li C. (GET, Toulouse, France), Longman J. (U. Oxford, UK), Martinez-Cortizas A. (U. Santiago de Compostela, Spain), Marx S. (U. Wollongong, Australia), Mattielli N. (U. Brussels, Belgium), Mighall T. (U. Aberdeen, UK), Nieminen T.M. (Natural Resources Institute, Helsinki, Finland), Piotrowska N. (GADAM Center, Poland), Pontevedra Pombal X. (U. Santiago de Compostela, Spain), Pratte S. (Zhejiang University), Renson V. (U. Missouri, USA), Shotyk W. (U. Alberta, Canada), Shuttleworth E. (U. Manchester, UK), Sikorski J. (GADAM Center, Poland), Talbot J. (U. Montreal, Canada), von Scheffer C. (U. Kiel, Germany), Weiss D. (Imperial College London, UK), Zaccone C. (U. Verona, Italy), Le Roux G. (CNRS, Toulouse, France)

Sincerely Yours,

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Reviewer #1: The authors have corrected the errors that I noted and have re-structured the paper so that it is improved substantially. The statements that are made are now factually correct. The remaining concern to me is the "tone" of the paper. While I appreciate that the authors' tone is less severe than in the previous version, at least in the abstract, the conclusion changes and once again it reads like an attack on the Miszazak et al paper. I strongly contend that if a paper is published primarily as a comment on another paper - the authors of the original paper should be allowed to comment. Is, as seems the case, the authors appear to be writing a paper that reiterates a scientific point that others are misinterpreting then I still feel it could be toned down without compromising this paper if the primary intent is to confirm that Pb is largely immobile in ombrotrophic peat and can reconstruct atmospheric Pb deposition. If it is the intent to criticize the Miszazak et al paper then it must go to these authors.

We thank the reviewer for his/her comment.

Let's not reverse things here. If anybody has been attacked our insulted, it is our community (hence we are so many to sign up this comment). It's not the opposite. This paper is appalling and should not have been published. While we can understand that STOTEN would not take the extreme decision of removing such flawed paper, we are left with the choice of putting things back. Hence there is indeed not much discussion. We merely recall the facts.

We have once again attempted to change the tone of our manuscript, although again, we were merely putting the facts, and strongly believed that yes, Miszczak et al failed to understand crucial concepts. There is no other explanation as the reviewer think there could be.

We however contest that the Miszczak et al. paper is deeply flawed and ignores a vast body of literature and for that deserves criticism. However, we agree it is important to ensure we focus on how Miszczak et al. interpret the data, but nevertheless think it is important to point out their key oversights.

About the comment: There seem to be a misunderstanding here. As we had already pointed out in our previous revised version, yes this is a comment. And yes, Miszczak et al. will have the right to reply to our comment, but once and only once our comment has passed the regular review process and is accepted for publication. Once our comment is accepted, STOTEN will made Miszczak et al. aware of it and allow them to reply. The STOTEN editor already confirmed us this process: Miszczak et al. will have the opportunity to reply. But of course they do not review our manuscript before publication.

My suggestions include:

1. Perhaps changing the title? If it is a comment on another paper those authors should be allowed to comment. As it in the review stage it will not compromise the publication.

We have not changed our title as it is indeed a comment, and Miszczak et al. will have the right to reply. We hope this is now clear with the explanations provided above. Such a title can be accepted (see past publications in STOTEN) and will appear in the "letter to the editor" section of a STOTEN volume, together with the reply of Miszczak et al.

We feel it is important to retain a direct reference to the Miszazak et al.'s paper as our contribution deals directly with the misapprehensions in that work.

2. The first sentence/abstract could simply be "Recent work has suggested that … In this paper we reiterate the evidence collected over 40 years of study that demonstrates Pb is largely immobile in ombrotrophic peat cores and highlight some errors that may lead to misinterpretation of this valuable archival record"

Changed as suggested.

3. The paper could be re-written slightly so that the points are retained but is not a direct criticism of this paper - for example.

Well, this is a direct criticism of that paper which is full of major flaws and that should not have been published… We have anyways adjusted the text.

4. Ombrotrophic peatlands are well-established archives of past atmospheric deposition of trace elements. After more than 40 years of investigation (see Table SI1), there is a consensus that Pb is largely immobile in ombrotrophic peatlands. We raise a series of critical issues regarding Miszczak et al. (2019) to clarify how ombrotrophic peatlands have allowed the reconstruction of past patterns of atmospheric metal contamination in the environment. We do not aim to provide a complete review of Pb in mires, however because the trophic status of a peatland is crucial to studies of past atmospheric metal deposition (see Supplementary Material for further details), we refer to a selection of studies relevant to the type of peatland that Miszczak et al. (2019) used, namely ombrotrophic peatlands, also known as bogs.

Could be written:

Ombrotrophic peatlands are well-established archives of past atmospheric deposition of trace elements. After more than 40 years of investigation (see Table SI1), there is a consensus that Pb is largely immobile in ombrotrophic peatlands. To avoid some confusion raised by a recent publication (Miszazak et al. 2019), we seek to clarify how ombrotrophic peatlands have allowed the reconstruction of past patterns of atmospheric metal contamination in the environment. We do not aim to provide a complete review of Pb in mires, however because the trophic status of a peatland is crucial to studies of past atmospheric metal deposition (see Supplementary Material for further details), we refer to a selection of studies relevant to ombrotrophic peatlands, also known as bogs.

We have re-phrased this section, although we don't use the Reviewer's exact wording, but have modified this section in a similar way.

5. I do not wish to go through the entire paper this way but hopefully my point is taken. The authors can still make all their valid points without continuously commenting on the Miszczak paper, but still make it clear that they had concerns with this paper. I would have no issues if the paper was written in this style. If it remains as presented, as a scientist myself, if someone where to directly attack my work I would want the chance to respond and I contend that it must go to the authors of the paper in question if that is the path chosen. For example, reading the sentence on line 106 is directly insulting the authors and makes an assumption that they "were unaware". Perhaps they are aware but had a different reason for doing what they did? On line 109, the authors claim "they failed

to comprehend" - this is derogatory terminology. My suggestions are easy to adopt: Section 2 is fine if it stops at line 47. In section 3, just delete the sentence beginning on line 60.

We have amended the manuscript as suggested throughout

6. Most changes are needed from sections 3.1 onwards, but they are simply editorial and are needed to change the tone and can easily be done, without compromising this paper in any way [I could likely do it in 30 minutes but it is not my paper]. I leave it to the authors to decide if they wish to do this but I do not recommend the paper be published unless they do. I do not wish to see reputable journals become a venue for arguments between research groups.

We take the Reviewers point and have amended our manuscript in light of these comments. This has necessitated some re-organisation of the manuscript to re-focus our comment on more explicitly on the utility of bogs and the necessary steps needed to reconstruct Pb contamination histories.

We feel it is important to also point out that we are not a research group, but are a group of authors representative of number of research groups working within this field. While a number of the authors have previously worked together, others have not. Nevertheless, when this paper appeared, we were all outraged by the low quality of the paper, the mis-interpretation of previous work (as well as some attacks), and yes, felt Miszczak et al. were insulting our community. We therefore felt the urge to write this comment. We however understand the reviewer as to that we should not reply by attacking or insulting, and have adjusted our manuscript once more, hopefully to the liking of the reviewer and editor.

Revised manuscript with changes marked
Click here to view linked References

Letter to the Editor - The utility of ombrotrophic peatlands for reconstructing metal contamination histories. Comment toon: "

A novel approach to peatlands as archives of total cumulative spatial pollution loads from atmospheric deposition of airborne elements complementary to EMEP data: priority pollutants (Pb, Cd, Hg)".

by Ewa Miszczak, Sebastian Stefaniak, Adam Michczyński, Eiliv Steinnes and Irena Twardowska.

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Abstract

A recent paper by Miszczak et al. (20192020); recently presented what they consider to be 'a A novel approach to using peatlands as archives of cumulative atmospheric pollution', Science of the Total Environment, 705, 135776) examines metal contamination in peat mires in Poland and Norway. The authors conclude that lead (Pb) records in ombrotrophic peatlands cannot be used to reconstruct the chronological history of anthropogenic activities due to post-depositional mobility of the metal. We contest this general conclusion which stands in contrast with a significant body of literature demonstrating that Pb is largely immobile in the vast majority of ombrotrophic peatlands. Our aim is to reaffirm the crucial contribution that peat bog records have made to our knowledge of atmospheric Pb pollution historiescontamination. ToIn addition, we re-iterate the necessity of following accepted protocols to produce reliable records We follow this bywe highlighting what we perceive are the most critical issues with the paper published by Miszczak et al. (2019)of anthropogenic Pb contamination in environmental archives.

Keywords

Lead, immobility, ombrotrophic peatland, bog, geochemistry, enrichment factor, metal accumulation rate

1. INTRODUCTION

Ombrotrophic peatlands are well-established archives of past atmospheric deposition of trace elements. After more than 40 years of investigation (see Table SI1), there is a consensus that Pb is largely immobile in ombrotrophic peatlands. We ~~argue~~contend that some of the conclusions reached ~~in a recent paper~~ by~~in~~ Miszczak et al (2020)~~19~~ are based on misinterpretation or incorrect ~~application of approaches to~~ sampl~~e collection~~ing and data analysis approaches. To avoid such confusion, we seek to clarify here how ombrotrophic peatlands have allowed the reconstruction of past patterns of atmospheric metal contamination in the environment ~~Correcting these misapprehensions is important because these techniques are well supported by evidence and are essential to producing reliable records and conclusions.~~. ~~We raise a series of critical issues regarding Miszczak et al. (2019) to clarify how ombrotrophic peatlands have allowed the reconstruction of past patterns of atmospheric metal contamination in the environment.~~ We however do not aim to provide a complete review of Pb in mires, ~~however~~ because the trophic status of a peatland is crucial to studies of past atmospheric metal deposition (see Supplementary Material for further details), we refer to a selection of studies relevant to ombrotrophic peatlands, also known as bogs. Due to their high atmospheric fidelity, ombrotrophic peatlands have special utility for reconstructing metal contamination records. This was also the type of peatland investigated by Miszczak et al. (2020).

~~the type of peatland that Miszczak et al. (2019) used, namely ombrotrophic peatlands, also known as bogs.~~

2. THE BEHAVIOUR OF LEAD IN OMBROTROPHIC PEATLANDS

While some studies have suggested that Pb can be mobilized in minerotrophic, riparian, drained or degraded peatlands (e.g. Syrovetnik et al., 2007; Smieja-Król et al., 2010, 2019; Rothwell, 2011; Broder and Biester, 2017) the majority of more than 40 years of literature suggests that Pb is largely immobile in ~~intact~~ pristine ombrotrophic peat profiles (e.g. De Vleeschouwer et al., 2010a; Marx et al., 2010, Shotyk et al., 2016a,b; Longman et al. 2018;

Fiałkiewicz-Kozieł et al., 2020 and references therein). Although Miszczak et al. (2020) cite literature to support their conclusion of Pb mobility in the bogs they examine, that literature pertains to minerotrophic or disturbed peatlands (e.g. Syrovetnik et al., 2007; Smieja-Król et al., 2010, 2019) and the processes that can promote Pb mobility in those systems are not applicable to ombrotrophic peatlands.

The similar elemental and isotopic trends encountered in ombrotrophic peat, lake sediment, ice and herbaria samples (e.g. Rosman et al., 1997; Weiss et al., 1999; Renberg et al., 2001; Farmer et al., 2002; Cloy et al. 2009; Bindler, 2011) and their agreement with anthropogenic emission~~s~~ patterns (e.g. Shotyk et al., 1998; Mighall et al., 2002; Kylander et al., 2006; De Vleeschouwer et al., 2009a; Marx et al., 2010; Bindler 2011; Cloy et al., 2008, 2009; Allan et al., 2013; Martínez-Cortizas et al., 2016) provide a body of evidence supporting the view that Pb is largely immobile in ~~ombrotrophic peatlands~~bogs. Significantly, stable Pb isotopes ~~in ombrotrophic~~records from ~~peat~~ bogs have consistently been found to accurately reflect temporal variability in source signatures in numerous studies (e.g. in reference *op. cit.*). This would not be the case if post-depositional mobility/isotope mixing ~~was~~ were taking place. Furthermore, in most ombrotrophic peat cores ^{210}Pb ages, as determined from the constant rate of supply (CRS) age-depth models (Appleby and Oldfield, 1978; Appleby, 2001), are in very good agreement with pollen chronological markers (Appleby et al., 1997), fallout radionuclide chronostratigraphic makers (e.g. from ^{14}C Bomb Pulse Curve, ^{137}Cs and ^{241}Am), and tephrochronology ~~in various locations~~ (e.g. Goodsite et al., 2001; Piotrowska et al. 2009; Li et al., 2017; Davies et al., 2018), providing *prima facie* evidence that ~~lead~~ Pb and its isotopes are largely immobile in bogs. Experimental studies lend further support to this (e.g. Vile et al., 1999; Novak et al., 2001). For example, Pb concentrations in the aqueous phase of ombrotrophic peatlands are low (<0.01% of total Pb), while the limited vertical water movement in bogs together with the size of the metal-containing particles in solution limits Pb redistribution (e.g. Shotyk et al., 2016b). Down-washing experiments have also demonstrated that Pb has limited mobility (Hansson et al. 2014, 2015).~~, while~~ ~~any~~ T~~t~~he limited mobility that may ~~occurred~~ occur ~~was~~ is not sufficient to compromise the use of Pb to reconstruct pollution historie~~y~~s over millennia. We note, however, ~~that~~ the spatial distribution of Pb ~~has to~~must be carefully addressed in cases where decomposition and compression integrate signals over longer (decadal and more) timespans (Bindler et al, 2004; Martinez Cortizas et al., 2012). Additionally, Pb behaviour in ombrotrophic peats has been demonstrated to differ from that of mobile elements such as Zn which, in contrast to Pb, displays evidence of vertical diffusion/advection as well as upward plant uptake (e.g. Shotyk 1988; Twardowska et al., 1999; Niemi~~n~~nen et al., 2002; Weiss et al., 2007).

In summary, there is a significant body of evidence demonstrating that Pb is largely immobile in bog profiles (see Table S1 and Supplementary Material) that stands in contrast to the conclusions of Miszczak et al. (2020).

~~Miszczak et al. (2019) fail to recognize the significant body of evidence demonstrating that Pb is largely immobile in bog profiles (see Table S1 and Supplementary Material). In addition, the use of it appears they were confused in thinking that literature pertaining to minerotrophic or disturbed peatlands (e.g. Syrovetnik et al., 2007; Smieja-Król et al., 2010, 2019) was to argue for Pb mobility in the bogs they examine is incorrect as the processes which can promote Pb mobility in these systems are not applicable to ombrotrophic peatlands, and thus incorrectly use that evidence to argue for Pb mobility in the bogs they examine.~~ .

3. HOW TO USE LEAD DATA TO ACCURATELY RECONSTRUCT HISTORICAL CONTAMINATION

In the following sections we outline what we consider to be the best practices to ensure accurate reconstruction of atmospheric Pb deposition. We also discuss appropriate approaches to ~~study~~ use Pb ~~contamination~~ pollution records ~~from~~ constructed from peatlands ~~as~~ to ~~records of~~examine contamin~~ate~~ant sources and to compare with emissions data. The approaches we outline are well established ~~not new~~ and have been described before (e.g. Givelet et al., 2004; De Vleeschouwer et al., 2010b). We hope that ~~these sections~~this overview corrects any misapprehensions arising from the incorrect application byapproaches used by Miszczak et al. (20192020). do not appear to be aware of theseof commonly applied peat sampling approaches and alsoas well as some misinterpretations previously published data.

3.1. Sampling and sub-sampling – Data resolution and geochronology

Correct coring and sub-sampling protocols are important tofor accurately reconstructing metal contamination deposition chronologiesrecords from peat mires. The slow accumulation rate of ombrotrophic peatlands means that peat sections oin the order of one vertical centimeter can represent decades of metal accumulation. For example, in European ombrotrophic peatlands, long-term mean peat accumulation rates (i.e. excluding surface vegetation growth) have been estimated to range betweenfrom c. 0.18 to 1 mm yr^{-1} (e.g. Gorham, 1991; Mäkilä, 1997; Malmer and Wallén, 2004; Pontevedra-Pombal, et al., 2017) depending on the vegetation and climate (e.g. Charman et al., 2013; Pontevedra-Pombal et al., 2019). Because of this, it is common-place for studies to both sample and date the living vegetation at the bog surface (e.g. Farmer et al., 2006; Kempter et al. 2007; Olid et al., 2008). This point was illustrated by Givelet et al. (2004) who stated: "*the historical record of atmospheric Pb … can depend to a large extent on the methods used to collect, handle, and prepare the samples for analysis*". As a result, high-resolution sub-sampling and dating are required to reconstruct decadal-scale atmospheric pollution records.

Here , including because the slow accumulation rate of peat means that peat sections can represent decades of deposition. Miszczak et al. (20192020) compare metal ncontaminants in their peat records to European Monitoring and Evaluation Program (EMEP) data (annual trace metal emissions, https://www.emep.int). Although potentially a very useful undertaking, . Unfortunately, their sampling approach unfortunately greatly reduces the usefulness utility of such atheir comparison. This is because Miszczak et al. (20192020) used followed the coring protocol of Steinnes and Sjøbakk (2005) where "*Sphagnum moss … and other plant material growing on the surface were removed…before the coring, and the reference surface* *level is thus the interface moss/peat. The thickness of the Sphagnum layer, if present, was always less than 10 cm".* In other words, the authors removed the living/surface vegetation which is an integral part of the ombrotrophic peat deposit, ~~and can~~potentially accumulating~~e~~ decades of information.

~~In other words, the authors removed the living/surface vegetation which is an integral part of the ombrotrophic peat deposit, and can accumulate decades of information.~~ Given the slow accumulation rates of bogs, the 40 years of EMEP data are likely, at best, to represent approximately 4 cm of peat accumulation (if surface vegetation is excluded and assuming a 1mm yr^{-1} peat accumulation rate). Therefore, the sampling resolution of Miszczak et al. (2020), where peats were subsampled in multi-centimeter increments, combined with their limited use of radionuclide dating, preclude any assessment of recent Pb deposition or comparison with EMEP data from their study. This is especially the case if surface vegetation were removed. ~~Th~~Slow growth rates, combined with the demonstrated importance of surface vegetation in accumulating metal contaminant~~te~~s means that Miszczak et al. (2020) assumption that ~~is approach, i.e., taking~~ the peat/vegetation interface ~~as representing~~represents the year of coring (in ~~this~~ that case 1999)~~,~~ is~~ more than likely~~ incorrect. Their approach ~~and~~~~would~~ therefore ~~-~~leads~~s~~ to large uncertainties ~~regarding any~~in chronology ~~or~~ and any inventory calculations performed thereafter. As previously stated, we consider comparing EMEP~~PA~~ data with data from ombrotrophic peatlands to be a very worthwhile undertaking, but it requires high-resolution sub-sampling and dating, which is unfortunately not achieved by Miszczak et al. (2020).~~.~~

~~The inappropriateness of the Miszczak et al. (2019) sampling approach is demonstrated by a large number of studies in which the living vegetation has been sampled for Pb and dated (e.g. Farmer et al., 2006; Kempter et al. 2007; Olid et al., 2008). This is further illustrated by Givelet et al. (2004) who state: "*the historical record of atmospheric Pb … can depend to a large extent on the methods used to collect, handle, and prepare the samples for analysis*". It is therefore unlikely that Miszczak et al. (2019) can accurately compare multi-centimeter~~

~~samples that represent several tens of years with EMEP data - itself only covering the last 40 years, especially if up to 10 cm of the uppermost vegetation (possibly representing up to 20 years of information) were not sampled. This would preclude both comparison with recent EMEP data and any evaluation of the reliability of peat bogs for reconstructing deposition chronologies.~~

~~If Miszczak et al. (2019) began sampling their peat record at the accumulating peat (i.e., ignoring the living vegetation), it is also unlikely that their sampling resolution would allow comparison with emissions data. In European ombrotrophic peatlands, long-term mean peat accumulation rates (i.e. excluding surface vegetation growth) have been estimated to range between c. 0.18 to 1 mm yr^{-1} (e.g. Gorham, 1991; Mäkilä, 1997; Malmer and Wallén, 2004; Pontevedra-Pombal, et al., 2017) depending on the vegetation and climate (e.g. Charman et al., 2013; Pontevedra-Pombal et al., 2019). As a result, high-resolution sub-sampling and dating are required to reconstruct decadal-scale atmospheric pollution records using peat cores and to properly compare them to 40 years of EMEP data (i.e. at best 4 cm of peat accumulation, surface vegetation excluded if we take a 1mm yr^{-1} peat accumulation rate). The low sub-sampling resolution of Miszczak et al. (2019) and their removal of surface vegetation, combined with limited radionuclide dating preclude any assessment of recent Pb deposition from their study.~~

3.2. Interpreting data in elemental ratios, enrichment factors (EFs) and metal accumulation rates,

Reconstructing Pb contamination in ombrotrophic peatlands requires an understanding of how ~~peat bogs~~they respond to environmental change. This is because changes in Pb concentrations may result from changes in the peat bog density/accumulation rate rather than changes in the extent of contamination. ~~- Miszczak et al. (2019~~2020) ~~seem~~ are perhaps unaware of th~~ese~~is important consideration~~s.~~ In the following section ~~which follows~~ we outline the importance of understanding density changes in peat records and the need to ~~take~~consider ~~into account~~the variability in natural Pb from aeolian mineral dust deposition.

We provide a brief overview of ~~They therefore misinterpret data published in De Vleeschouwer et al. (2009a) and instead assume that those authors incorrectly interpreted their own data. Consequently, Miszczak et al. (2019) appear to misinterpret or fail to comprehend: 1) the effect of the density record presented in De Vleeschouwer et al. (2009b) and why the density and accumulation rates change within the Slowinskie Blota peat profile, and 2)~~ techniques to account for such changes, allowing ~~and to reliably use~~ Pb to be reliably used as a tracer of past anthropogenic activity.

3.2.1. Density, accumulation rate and Pb concentration

~~Within~~ The importance of understanding and accounting for changes in peat ~~in~~ density and dust input are illustrated by De Vleeschouwer et al.~~,~~ (2009a,b) in their study of the Slowinskie Blota ombrotrophic peatland ~~in~~ (Poland). ~~That peatland is located close to those examined by Miszczak et al. (2020).~~ In ~~the the Slowinskie Blota~~this peatland ~~core~~, a section of higher than average bulk density was present between 50 and 35 cm depth (De Vleeschouwer et al., 2009a,b). It corresponds to the timing of the Little Ice Age, when colder temperatures promoted a decrease in peat accumulation rates coeval with increased windiness and dune activity (i.e. increased aeolian lithogenic inputs~~; De Vleeschouwer et al., 2009b~~). This combination of reduced organic accumulation rates and increased dust input resulted in ~~in the~~an increase in bulk density~~ in the bog~~. The ~~result on the~~effect of these changes ~~Pb concentration profile~~ was an ~~apparent~~ increase in Pb concentration within the peat profile. Although part of this Pb increase is attributable to increased pollution in the Industrial Revolution, the majority of the Pb increase results from the decrease in peat accumulation causing an apparent increase in pollution Pb ~~contamination~~ accumulation. This occurs because that section of the peat profile represents a greater period of time ~~by comparison~~than ~~the~~ sections below 50 cm depth or above 35 cm depth. In addition, increased dust inputs during the drier conditions of the Little Ice Age mean there was an increase in natural Pb input during that period. Miszczak et al. (2020) ~~incorrectly~~ incorrectly assumed the increase in Pb in the Slowinskie Blota peatland at that time resulted from the movement of Pb from higher in the peat profile (i.e., Pb mobility in the bog). But, by appropriately accounting for the change in density and increased dust input during the Little Ice Age (i.e. using EFs accumulation rates and isotopic ratios, which are discussed in the next section), De Vleeschouwer et al.~~,~~ (2009a~~,b~~) demonstrated the maximum Pb concentration in the Slowinskie Blota record occurred at AD 1960-70s, and not between 50 and 35 cm depth as the raw Pb concentration data would suggest. ~~Thus~~The maximum Pb contamination therefore coincided precisely with maximum Pb emissions from leaded gasoline, just prior to Pb being banned and phased out beginning in the 1980s (e.g. Pacyna and Pacyna, 2000). De Vleeschouwer et al.~~,~~ (2009a,b)~~ therefore showed that the Pb contamination record in Slowinskie Blota ~~, matches the known history of anthropogenic Pb emissions in Europe clearly demonstrating that it is not related to any post-depositional mobility.~~,~~ The approach to accurately reconstruct Pb contamination (as separate from total Pb concentrations) is outlined in the following section.

~~owing largely to the decrease in peat accumulation in addition to displaying an increase in the concentrations of pollutant elements from the Industrial Revolution. Indeed, the maximum Pb concentration in the Slowinskie Blota record occurred at AD 1960-70s, coinciding with maximum Pb emissions from leaded gasoline, just prior to Pb being banned and phased out beginning in the 1980s (e.g. Pacyna and Pacyna, 2000). It has therefore nothing to do with any post-depositional Pb migration as claimed by Miszczak et al. (2019).~~

3.2.2. Pb as a tracer of past anthropogenic activity

Since the pioneering paper of Lee and Tallis (1973), practices have developed to ensure the accurate use of trace metal data to reconstruct past environmental pollution. It has been demonstrated that using concentration data to reconstruct past anthropogenic activity is problematic because, as shown above, peat accumulation ~~and accumulation~~ rates alter total Pb concentrations~~alter total Pb concentrations~~. A~~.~~ Additionally, because the rate of dust deposition in bogs (from wind erosion of soils) varies~~ in time~~, it is necessary to separate natural Pb in dust from anthropogenic Pb~~ within any studied bog sediments~~. ~~using EFs~~

~~allows the Pb from anthropogenic contamination to be separated from Pb contained in natural dust deposited in the bog from wind erosion of soils.~~Therefore, it is common practice to use metal to lithogenic element ratios, enrichment factors (EFs), or elemental mass accumulation rates (e.g. Shotyk et al., 1998; Le Roux et al., 2010; Allan et al., 2013) to reconstruct contamination histories. These approaches are important to avoid~~ing~~ misinterpretations based on examining concentration data alone.~~.~~ ~~Additionally, using EFs allows the Pb from anthropogenic contamination to be separated from Pb contained in natural dust deposited in the bog from wind erosion of soils.~~ ~~Unfortunately,~~ Miszczak et al. (~~2019~~2020) did not apply ~~one of~~ these standard approaches. ~~and, as a result, draw erroneous conclusions. They misinterpret the concentration data~~ By comparison De Vleeschouwer et al. (2009a) use EFs and Pb accumulation rates (combined ~~with~~to ~~use of~~ Pb isotopes and high-resolution sampling and dating including ~~of~~ the surface vegetation) to come to a different set of conclusions regarding the utility of Polish ombrotrophic peatlands for reconstructing Pb contamination histories. Consequently ~~they provide~~the latter represents a more accurate picture of the extent of Pb contamination in Poland over the past 1400 years, demonstrating *i)* ~~presented De Vleeschouwer et al. (2009a), who by contrast, use EFs and Pb accumulation rates (combined with high-resolution sampling and dating including the surface vegetation) to provide a more accurate picture of the extent of Pb contamination in Poland over the past 1400 years. Importantly, De Vleeschouwer et al. (2009a) demonstrate that~~ the Pb accumulation rate in the topmost centimeter of the peat ~~core~~ is of the same order of magnitude as ~~the that of the~~ 2009 European Pb deposition (www.msceast.org, www.emep.int), *ii)* the main sources of anthropogenic Pb are from metallurgy, coal and gasoline and, *iii)* the peak in Pb contamination matches the history of European Pb emissions~~. Their findings are further supported by Pb isotope data, which confirm the history of anthropogenic Pb by identifying the sources of Pb in Poland (metallurgy, coal, gasoline) and demonstrating the immobility of Pb in this case~~. That work therefore provides another example~~,~~ amongst many others, (Table

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SI1) ~~demonstrating that~~of Pb ~~is~~ being largely immobile in ombrotrophic peatlands and show~~s~~ing peatlands to be excellent recorders of anthropogenic activities.

4. CONCLUSIONS

Due to their ~~atmospheric~~ fidelity ombrotrophic peatlands have been extensively used to study current and past patterns of atmospheric metal contamination and metal use, in particular for Pb. Despite their utility there are some key considerations required when constructing contamination histories from ~~peat~~ bogs. The aim of this comment was to highlight some these considerations. The impetus for this arose from the recent paper by Miszczak et al (2020) who used nonstandard sampling and analysis techniques and, as a result, came to what we consider to be erroneous conclusions. Additional discussion on the effect of pH on Pb mobility and the relationship between peat age and the history of Pb contamination is provided in the supplementary material.

Over the past 40 years ~~a large number of~~many investigators have developed or applied a range of analysis and sampling techniques necessary to construct metal contamination records in ombrotrophic peatlands. These approaches include undertaking high resolution sampling and dating, including sampling the living vegetation of~~n~~ the surface of ~~peat~~ bogs and the use of short-~~-~~lived radionuclides (such as ^{210}Pb) to accurately reconstruct metal contamination over the past ~100 years or less. They also include calculating enrichment factors (EFs), elemental ratios, or using accumulations rates (as opposed to raw metal concentration data) to take account of changes in peat density/growth rates and changes in natural metal input. Studies of Pb contamination have also benefitted from the use of Pb isotopes to decipher emission sources at a regional to continental scale. Many of these steps are also necessary when accurately determining contaminate loads and patterns in other environments including in ice, lakes and soils and within direct atmospheric samples~~,~~. ~~a~~Although. W~~w~~e therefore wish to reiterate the particular value of ombrotrophic peats for reconstructing atmospheric metal contamina~~tion~~nt chronologies due to their wide distribution and high ~~atmospheric~~ fidelity. We maintain that 40 years of literature demonstrate that Pb is

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largely immobile in ombrotrophic peatlands (i.e. bogs) and that peat cores extracted from th~~ese~~is type of mire~~s~~ represent reliable archives for reconstructing past natural changes in Pb deposition from natural processes and anthropogenic activity. The approach is supported by experimental work and similar reconstructions of metal contamination in other environmental archives (herbarium samples, lake sediments, ice cores). We conclude by noting that reconstructions of Pb contamination from ~~peat~~ bogs provide unequivocal ~~records~~evidence of ~~of changes in~~ the global -scale of -atmospheric Pb contamination and a reliable record of the timing of changes in atmospheric deposition extending from pre-history until the present day.~~.~~

~~What~~

~~we perceive as a failure of Miszczak et al. (2019) to fully appreciate appropriate and well described peat sampling and data analysis practices provided the impetus for us to write this comment. Unfortunately, Miszczak et al. (2019) misinterpreted both their data and existing literature, leading to erroneous conclusions. Our comment attempts to highlight some key considerations required when constructing pollution histories from peat bogs. We also note there are a number of other issues with the paper of Miszczak et al. (2019) that are not addressed here, including some unsupported assumptions on the effect of pH on Pb mobility or the relationship between peat age and the history of Pb contamination, a number of which are addressed further in supplementary material. We strongly disagree with conclusion of Miszczak et al. (2019) that Pb deposition chronologies cannot be reconstructed due to bulk density-controlled element vertical redistribution, i.e. Pb mobility within ombrotrophic peat. We maintain that 40 years of literature demonstrate that Pb is largely immobile in ombrotrophic peatlands (i.e. bogs) and that peat cores extracted from these mires represent reliable archives for reconstructing past natural changes in Pb deposition as well as inputs from anthropogenic activity. Coupling elemental ratios, enrichment factors and Pb isotopes to high-resolution sampling and accurate age dating allows deciphering of emission sources at a regional to continental scale. The approach is supported by experimental works and similar~~

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~~reconstructions of metal contamination in other environmental archives (herbarium samples, lake sediments, ice cores). We conclude by noting that reconstructions of Pb contamination from peat bogs provide unequivocal records of changes in global atmospheric Pb contamination.~~

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Comment on: "A novel approach to peatlands as archives of total cumulative spatial pollution loads from atmospheric deposition of airborne elements complementary to EMEP data: priority pollutants (Pb, Cd, Hg)"

by Ewa Miszczak, Sebastian Stefaniak, Adam Michczyński, Eiliv Steinnes and Irena Twardowska.

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Abstract

A recent paper by Miszczak et al. (2020) examines metal contamination in mires in Poland and Norway. The authors conclude that lead (Pb) records in ombrotrophic peatlands cannot be used to reconstruct the chronological history of anthropogenic activities due to post-depositional mobility of the metal. We contest this general conclusion which stands in contrast with a significant body of literature demonstrating that Pb is largely immobile in the vast majority of ombrotrophic peatlands. Our aim is to reaffirm the crucial contribution that peat records have made to our knowledge of atmospheric Pb contamination. In addition, we re-iterate the necessity of following accepted protocols to produce reliable records of anthropogenic Pb contamination in environmental archives.

Keywords

Lead, immobility, ombrotrophic peatland, bog, geochemistry, enrichment factor, metal accumulation rate

1. INTRODUCTION

Ombrotrophic peatlands are well-established archives of past atmospheric deposition of trace elements. After more than 40 years of investigation (see Table SI1), there is a consensus that Pb is largely immobile in ombrotrophic peatlands. We contend that some of the conclusions reached by Miszczak et al (2020) are based on misinterpretation or incorrect sampling and data analysis approaches. To avoid such confusion, we seek to clarify here how ombrotrophic peatlands have allowed the reconstruction of past patterns of atmospheric metal contamination in the environment. We however do not aim to provide a complete review of Pb in mires, because the trophic status of a peatland is crucial to studies of past atmospheric metal deposition (see Supplementary Material for further details), we refer to a selection of studies relevant to ombrotrophic peatlands, also known as bogs. Due to their high atmospheric fidelity, ombrotrophic peatlands have special utility for reconstructing metal contamination records. This was also the type of peatland investigated by Miszczak et al. (2020).

2. THE BEHAVIOUR OF LEAD IN OMBROTROPHIC PEATLANDS

While some studies have suggested that Pb can be mobilized in minerotrophic, riparian, drained or degraded peatlands (e.g. Syrovetnik et al., 2007; Smieja-Król et al., 2010, 2019; Rothwell, 2011; Broder and Biester, 2017) the majority of more than 40 years of literature suggests that Pb is largely immobile in pristine ombrotrophic peat profiles (e.g. De Vleeschouwer et al., 2010a; Marx et al., 2010, Shotyk et al., 2016a,b; Longman et al. 2018; Fiałkiewicz-Kozieł et al., 2020 and references therein). Although Miszczak et al. (2020) cite literature to support their conclusion of Pb mobility in the bogs they examine, that literature pertains to minerotrophic or disturbed peatlands (e.g. Syrovetnik et al., 2007; Smieja-Król et al., 2010, 2019) and the processes that can promote Pb mobility in those systems are not applicable to ombrotrophic peatlands.

The similar elemental and isotopic trends encountered in ombrotrophic peat, lake sediment, ice and herbaria samples (e.g. Rosman et al., 1997; Weiss et al., 1999; Renberg et al., 2001; Farmer et al., 2002; Cloy et al. 2009; Bindler, 2011) and their agreement with anthropogenic emission patterns (e.g. Shotyk et al., 1998; Mighall et al., 2002; Kylander et al., 2006; De Vleeschouwer et al., 2009a; Marx et al., 2010; Bindler 2011; Cloy et al., 2008, 2009; Allan et al., 2013; Martínez-Cortizas et al., 2016) provide a body of evidence supporting the view that Pb is largely immobile in bogs. Significantly, stable Pb isotopes records from bogs have consistently been found to accurately reflect temporal variability in source signatures in numerous studies (e.g. in reference *op. cit.*). This would not be the case if post-depositional mobility/isotope mixing were taking place. Furthermore, in most ombrotrophic peat cores ^{210}Pb ages, as determined from the constant rate of supply (CRS) age-depth models (Appleby and Oldfield, 1978; Appleby, 2001), are in very good agreement with pollen chronological markers (Appleby et al., 1997), fallout radionuclide chronostratigraphic makers (e.g. from ^{14}C Bomb Pulse Curve, ^{137}Cs and ^{241}Am), and tephrochronology (e.g. Goodsite et al., 2001; Piotrowska et al. 2009; Li et al., 2017; Davies et al., 2018), providing *prima facie* evidence that Pb and its isotopes are largely immobile in bogs. Experimental studies lend further support to this (e.g. Vile et al., 1999; Novak et al., 2001). For example, Pb concentrations in the aqueous phase of ombrotrophic peatlands are low (<0.01% of total Pb), while the limited vertical water movement in bogs together with the size of the metal-containing particles in solution limits Pb redistribution (e.g. Shotyk et al., 2016b). Down-washing experiments have also demonstrated that Pb has limited mobility (Hansson et al. 2014, 2015). The limited mobility that may occur is not sufficient to compromise the use of Pb to reconstruct pollution histories over millennia. We note, however, the spatial distribution of Pb must be carefully addressed in cases where decomposition and compression integrate signals over longer (decadal and more) timespans (Bindler et al, 2004; Martinez Cortizas et al., 2012). Additionally, Pb behaviour in ombrotrophic peats has been demonstrated to differ from that of mobile elements such as Zn which, in contrast to Pb, displays evidence of vertical diffusion/advection as well as upward plant uptake (e.g. Shotyk 1988; Twardowska et al., 1999; Nieminen et al., 2002; Weiss et al., 2007).

In summary, there is a significant body of evidence demonstrating that Pb is largely immobile in bog profiles (see Table S1 and Supplementary Material) that stands in contrast to the conclusions of Miszczak et al. (2020).

3. HOW TO USE LEAD DATA TO ACCURATELY RECONSTRUCT HISTORICAL CONTAMINATION

In the following sections we outline what we consider to be the best practices to ensure accurate reconstruction of atmospheric Pb deposition. We also discuss appropriate approaches to use Pb pollution records constructed from peatlands to examine contaminant sources and to compare with emissions data. The approaches we outline are well established and have been described before (e.g. Givelet et al., 2004; De Vleeschouwer et al., 2010b). We hope that this overview corrects any misapprehensions arising from the approaches used by Miszczak et al. (2020).

3.1. Sampling and sub-sampling – Data resolution and geochronology

Correct coring and sub-sampling protocols are important for accurately reconstructing metal contamination records from mires. The slow accumulation rate of ombrotrophic peatlands means that peat sections on the order of one vertical centimeter can represent decades of metal accumulation. For example, in European ombrotrophic peatlands, long-term mean peat accumulation rates (i.e. excluding surface vegetation growth) have been estimated to range from c. 0.18 to 1 mm yr^{-1} (e.g. Gorham, 1991; Mäkilä, 1997; Malmer and Wallén, 2004; Pontevedra-Pombal, et al., 2017) depending on the vegetation and climate (e.g. Charman et al., 2013; Pontevedra-Pombal et al., 2019). Because of this, it is commonplace for studies to both sample and date the living vegetation at the bog surface (e.g. Farmer et al., 2006; Kempter et al. 2007; Olid et al., 2008). This point was illustrated by Givelet et al. (2004) who stated: "*the historical record of atmospheric Pb … can depend to a large extent on the* *methods used to collect, handle, and prepare the samples for analysis*". As a result, high-resolution sub-sampling and dating are required to reconstruct decadal-scale atmospheric pollution records.

Here Miszczak et al. (2020) compare metal contaminants in their peat records to European Monitoring and Evaluation Program (EMEP) data (annual trace metal emissions, https://www.emep.int). Although potentially a very useful undertaking, their sampling approach unfortunately greatly reduces the utility of their comparison. This is because Miszczak et al. (2020) followed the coring protocol of Steinnes and Sjøbakk (2005) where "*Sphagnum moss … and other plant material growing on the surface were removed…before the coring, and the reference surface level is thus the interface moss/peat. The thickness of the Sphagnum layer, if present, was always less than 10 cm".* In other words, the authors removed the living/surface vegetation which is an integral part of the ombrotrophic peat deposit, potentially accumulating decades of information.

Given the slow accumulation rates of bogs, the 40 years of EMEP data are likely, at best, to represent approximately 4 cm of peat accumulation (if surface vegetation is excluded and assuming a 1mm yr^{-1} peat accumulation rate). Therefore, the sampling resolution of Miszczak et al. (2020), where peats were subsampled in multi-centimeter increments, combined with their limited use of radionuclide dating, preclude any assessment of recent Pb deposition or comparison with EMEP data from their study. This is especially the case if surface vegetation were removed. Slow growth rates, combined with the demonstrated importance of surface vegetation in accumulating metal contaminants means that Miszczak et al. (2020) assumption that the peat/vegetation interface represents the year of coring (in that case 1999) is incorrect. Their approach therefore leads to large uncertainties in chronology and any inventory calculations performed thereafter. As previously stated, we consider comparing EMEP data with data from ombrotrophic peatlands to be a very worthwhile undertaking, but it requires high-resolution sub-sampling and dating, which is unfortunately not achieved by Miszczak et al. (2020).

3.2. Interpreting data in elemental ratios, enrichment factors (EFs) and metal accumulation rates,

Reconstructing Pb contamination in ombrotrophic peatlands requires an understanding of how they respond to environmental change. This is because changes in Pb concentrations may result from changes in the peat bog density/accumulation rate rather than changes in the extent of contamination. In the following section we outline the importance of understanding density changes in peat records and the need to consider the variability in natural Pb from aeolian mineral dust deposition. We provide a brief overview of techniques to account for such changes, allowing Pb to be reliably used as a tracer of past anthropogenic activity.

3.2.1. Density, accumulation rate and Pb concentration

The importance of understanding and accounting for changes in peat density and dust input are illustrated by De Vleeschouwer et al. (2009a,b) in their study of the Slowinskie Blota ombrotrophic peatland (Poland). In this peatland, a section of higher than average bulk density was present between 50 and 35 cm depth (De Vleeschouwer et al., 2009a,b). It corresponds to the timing of the Little Ice Age, when colder temperatures promoted a decrease in peat accumulation rates coeval with increased windiness and dune activity (i.e. increased aeolian lithogenic inputs). This combination of reduced organic accumulation rates and increased dust input resulted in an increase in bulk density. The effect of these changes was an increase in Pb concentration within the peat profile. Although part of this Pb increase is attributable to increased pollution in the Industrial Revolution, the majority of the Pb increase results from the decrease in peat accumulation causing an apparent increase in pollution Pb accumulation. This occurs because that section of the peat profile represents a greater period of time than sections below 50 cm depth or above 35 cm depth. In addition, increased dust inputs during the drier conditions of the Little Ice Age mean there was an increase in natural Pb input during that period. Miszczak et al. (2020) incorrectly assumed the increase in Pb in the Slowinskie Blota peatland at that time resulted from the movement of Pb from higher in the peat profile (i.e., Pb mobility in the bog). But, by appropriately accounting for the change in density and increased dust input during the Little Ice Age (i.e. using EFs accumulation rates and isotopic ratios, which are discussed in the next section), De Vleeschouwer et al. (2009a) demonstrated the maximum Pb concentration in the Slowinskie Blota record occurred at AD 1960-70s, and not between 50 and 35 cm depth as the raw Pb concentration data would suggest. The maximum Pb contamination therefore coincided precisely with maximum Pb emissions from leaded gasoline, just prior to Pb being banned and phased out beginning in the 1980s (e.g. Pacyna and Pacyna, 2000). De Vleeschouwer et al. (2009a,b), match the known history of anthropogenic Pb emissions in Europe clearly demonstrating that it is not related to any post-depositional mobility. The approach to accurately reconstruct Pb contamination (as separate from total Pb concentrations) is outlined in the following section.

3.2.2. Pb as a tracer of past anthropogenic activity

Since the pioneering paper of Lee and Tallis (1973), practices have developed to ensure the accurate use of trace metal data to reconstruct past environmental pollution. It has been demonstrated that using concentration data to reconstruct past anthropogenic activity is problematic because, as shown above, peat accumulation rates alter total Pb concentrations. Additionally, because the rate of dust deposition in bogs (from wind erosion of soils) varies, it is necessary to separate natural Pb in dust from anthropogenic Pb. Therefore, it is common practice to use metal to lithogenic element ratios, enrichment factors (EFs), or elemental mass accumulation rates (e.g. Shotyk et al., 1998; Le Roux et al., 2010; Allan et al., 2013) to reconstruct contamination histories. These approaches are important to avoid misinterpretations based on examining concentration data alone. Miszczak et al. (2020) did not apply these standard approaches. By comparison De Vleeschouwer et al. (2009a) use EFs and Pb accumulation rates (combined to Pb isotopes and high-resolution sampling and dating including the surface vegetation) to come to a different set of conclusions regarding the utility of Polish ombrotrophic peatlands for reconstructing Pb contamination histories.

Consequently the latter represents a more accurate picture of the extent of Pb contamination in Poland over the past 1400 years, demonstrating *i)* the Pb accumulation rate in the topmost centimeter of the peat is of the same order of magnitude as 2009 European Pb deposition (www.msceast.org, www.emep.int), *ii)* the main sources of anthropogenic Pb are from metallurgy, coal and gasoline and, *iii)* the peak in Pb contamination matches the history of European Pb emissions. That work therefore provides another example amongst many others, (Table SI1) of Pb being largely immobile in ombrotrophic peatlands and shows peatlands to be excellent recorders of anthropogenic activities.

4. CONCLUSIONS

Due to their fidelity ombrotrophic peatlands have been extensively used to study current and past patterns of atmospheric metal contamination and metal use, in particular for Pb. Despite their utility there are some key considerations required when constructing contamination histories from bogs. The aim of this comment was to highlight some these considerations. The impetus for this arose from the recent paper by Miszczak et al (2020) who used nonstandard sampling and analysis techniques and, as a result, came to what we consider to be erroneous conclusions. Additional discussion on the effect of pH on Pb mobility and the relationship between peat age and the history of Pb contamination is provided in the supplementary material. Over the past 40 years many investigators have developed or applied a range of analysis and sampling techniques necessary to construct metal contamination records in ombrotrophic peatlands. These approaches include undertaking high resolution sampling and dating, including sampling the living vegetation of the surface of bogs and the use of short-lived radionuclides (such as ^{210}Pb) to accurately reconstruct metal contamination over the past ~100 years or less. They also include calculating enrichment factors (EFs), elemental ratios, or using accumulations rates (as opposed to raw metal concentration data) to take account of changes in peat density/growth rates and changes in natural metal input. Studies of Pb contamination have also benefitted from the use of Pb isotopes to decipher emission sources at a regional to continental scale. Many of these steps are also necessary when accurately determining contaminate loads and patterns in other environments including in ice, lakes and soils and within direct atmospheric samples. We therefore wish to reiterate the particular value of ombrotrophic peats for reconstructing atmospheric metal contaminant chronologies due to their wide distribution and high fidelity. We maintain that 40 years of literature demonstrate that Pb is largely immobile in ombrotrophic peatlands (i.e. bogs) and that peat cores extracted from this type of mire represent reliable archives for reconstructing past natural changes in Pb deposition from natural processes and anthropogenic activity. The approach is supported by experimental work and similar reconstructions of metal contamination in other environmental archives (herbarium samples, lake sediments, ice cores). We conclude by noting that reconstructions of Pb contamination from bogs provide unequivocal evidence of the global scale of atmospheric Pb contamination and a reliable record of the timing of changes in atmospheric deposition extending from pre-history until the present day.

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*conflict of Interest Statement

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Subject: Conflict of Interest Statement

Buenos Aires, March 13th, 2020

To Science of the Total Environment Co-Editors in chief Damià Barceló and Jay Gan.

Dear Dr Barcelo and Dr Gan,

On Behalf of all my co-authors, I hereby declare I have no conflict of interest with any of the authors of Miszczak et al. (2019).

On the behalf of: Baron S. (CNRS, Toulouse, France), Cloy J.M. (Scotland's Rural College, Edinburgh, UK), Enrico M. (Harvard, USA), Ettler V. (Charles University Prague, Czech Republic), Fagel N. (U. Liège, Belgium), Kempter H., Kylander M. (U. Stockholm, Sweden), Li C. (GET, Toulouse, France), Longman J. (U. Oxford, UK), Martinez-Cortizas A. (U. Santiago de Compostela, Spain), Marx S. (U. Wollongong, Australia), Mattielli N. (U. Brussels, Belgium), Mighall T. (U. Aberdeen, UK), Nieminen T.M. (Natural Resources Institute, Helsinki, Finland), Piotrowska N. (GADAM Center, Poland), Pontevedra Pombal X. (U. Santiago de Compostela, Spain), Pratte S. (Zhejiang University), Renson V. (U. Missouri, USA), Shotyk W. (U. Alberta, Canada), Shuttleworth E. (U. Manchester, UK), Sikorski J. (GADAM Center, Poland), Talbot J. (U. Montreal, Canada), von Scheffer C. (U. Kiel, Germany), Weiss D. (Imperial College London, UK), Zaccone C. (U. Verona, Italy), Le Roux G. (CNRS, Toulouse, France)

Sincerely Yours,

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Supplementary Material

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Supplementary Table

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