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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

François de Vleeschouwer, S. Baron, J.M. Cloy, M. Enrico, V. Ettler, N. Fagel, H. Kempter, M. Kylander, C. Li, J. Longman, et al.

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Letter to the Editor - Comment to:

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 Michzynski, Eiliv Steinnes and Irena Twardowska.

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Abstract

Miszczak et al. (2019) recently published a dataset in *Science of the Total Environment* presenting it as a novel approach to using peatlands as archives of atmospheric pollution. Problematically, the authors claim that, because of possible post-depositional mobility, lead (Pb) records in ombrotrophic peatlands cannot be used to reconstruct the chronological history of anthropogenic activities. While for mercury (Hg) there are some open questions (e.g. Biester et al. 2007; Enrico et al., 2016), and whereas cadmium (Cd) has not been studied with much detail (e.g. McBride et al., 2005), Pb has been extensively studied. Here, referring to decades of literature in the field, we summarize the evidence for the immobility of this element in ombrotrophic peatlands, and reaffirm the crucial contribution of peat records to our knowledge of past Pb atmospheric circulation and deposition.

Keywords

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

1. INTRODUCTION

Ombrotrophic peatlands are excellent archives to study the atmospheric deposition of many trace elements. Most studies to date have focused on two areas: (1) the reconstruction of past trace metals pollution and (2) assessing trace metal mobility. The observation of a distinct Pb peak dating from Roman times in ombrotrophic peatlands from England (Lee and Tallis, 1973) and the determination of modern high Pb concentrations in Scandinavian bogs (Hvatum et al., 1983) were the reasons why Pb received most of the attention in recent decades. These initial findings were corroborated by the existence of a common pattern of atmospheric Pb deposition throughout the Northern Hemisphere, from Sweden (Brännvall et al., 1997; Klaminder et al., 2003), Switzerland (Shotyk et al., 1999; Weiss et al., 1999), the United Kingdom (Farmer et al., 1997; West et al., 1997; Le Roux et al., 2004) and Spain (Martínez Cortizas et al., 2002), to cite only a few. Such studies mainly focused on ombrotrophic peatlands (i.e. bogs), which reliably record changes in long-term atmospheric pollution (e.g. Shotyk et al., 1998, Kempter and Frenzel, 1999, Martinez-Cortizas et al., 2002; Le Roux et al., 2005; Aubert, 2006; Coggins, 2006; Weiss et al., 2007, Kylander et al., 2009, De Vleeschouwer et al., 2009a, 2014; Allan et al., 2013, Longman et al., 2018). Other studies have utilized minerotrophic peatlands (i.e. fens) where ombrotrophic peatlands were not available (Shotyk, 2002; Sonke et al., 2002; Monna et al., 2004; Baron et al., 2005). In some cases, even when fen peat does not solely record atmospheric inputs, pollution can be detected, especially if a dominant pollution source is in the vicinity of the studied site (West et al., 1997; Mighall et al., 2002; Baron et al., 2005, Renson et al., 2008). Other studies have focused on the record of pre-industrial geochemical signature of peat (e.g. Martinez-Cortizas et al., 1999; Shotyk et al., 2001, Martinez-Cortizas et al., 2002; Roos-Barraclough et al., 2002; Weiss et al., 2002; Kylander et al., 2005, 2007). The second area of investigation - metal mobility in peat - has been developed because the behavior of trace metals is linked to various phenomena such as adsorption onto organic matter (e.g. Bunzl et al., 1976; Grybos et al., 2007) and scavenging by oxy-hydroxides (e.g. Tessier et al., 1996).

29 After more than 40 years of investigation, there is a consensus about the vertical immobility
30 of Pb in ombrotrophic peatlands based on i) the coherence and synchronicity of the Pb
31 pollution patterns identified in ombrotrophic peatlands over a wide latitudinal distribution; ii) a
32 constant improvement of the age-depth models; iii) the improvement of analytical techniques
33 and statistics; iv) the generalization of multiproxy studies. This comment aims at clarifying
34 key critical issues in Miszczak et al. (2019), to help avoid reader confusion and to clarify how
35 ombrotrophic peatland geochemistry help reconstructing past metal contamination in the
36 environment.

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39 2. **LEAD MOBILITY**

40 Whilst a small number of papers have suggested Pb mobility under particular settings
41 (Rauch et al. 2005b, Gaca et al., 2006), or discussed metal mobility in minerotrophic or
42 drained peatlands (Syrovetsnik et al., 2007; Smieja-Król et al., 2010, 2019), the majority of
43 more than 40 years of literature indicates Pb is immobile in ombrotrophic peat. This has been
44 demonstrated through various experiments summarized here as well as in in a number of
45 review papers (e.g. Le Roux et al., 2005; De Vleeschouwer et al., 2010a; Hansson et al.,
46 2015; Shotyk et al., 2016a). Miszczak et al. (2019) however ignore this large body of
47 evidence. We list below a series of evidence supported by Table 1 that provide the main
48 references in the field, to argue that Pb is indeed immobile in ombrotrophic peat.

49

50 A. ***Regional or continental comparison of lead profiles in ombrotrophic peatlands,***
51 ***lake sediments and ice cores provide similar trends*** (e.g. Rosman et al., 1997;
52 Renberg et al., 2001; De Vleeschouwer et al., 2010a and ref. therein; Bindler, 2011;
53 Hansson et al., 2015a) and match the history of regional anthropogenic lead use, such
54 as mining and metallurgy (e.g. Shotyk et al., 1998; Kempter and Frenzel, 1999; Bindler
55 et al., 2011; Allan et al., 2013) or emission data (e.g. Le Roux et al., 2006; De
56 Vleeschouwer et al., 2009a; Cloy et al., 2011; Allan et al., 2013). Furthermore, when

57 ombrotrophic peatland records are compared to nearby varved lakes (e.g. Renberg et
58 al., 2001; Bindler, 2011; Hansson et al., 2015a), profiles are similar, demonstrating that
59 both archives can reliably reconstruct past Pb contamination on a continental scale, and
60 that Pb is immobile in ombrotrophic peat over millennial timescales.

61 **B. *Lead immobility was demonstrated by comparing ombrotrophic peat cores and***
62 ***herbarium Sphagnum samples*** collected at the exact same sites each year for more
63 than 150 years (Weiss et al., 1999; Farmer et al., 2002; Shotbolt et al. 2007). This
64 evidence is independent from any post-depositional process. The Pb pollution trends
65 reconstructed from the peat core and the herbarium samples are very similar, once
66 again providing compelling evidence of Pb immobility in ombrotrophic peatlands. Such
67 cumulative evidence of all the different media (peat, sediment, ice, herbaria) for the past
68 150 years shows cohesion that is a strong argument against any subsequent post-
69 depositional migration (Bindler et al., 2011).

70 **C. *Lead isotopes have also provided strong evidence of Pb immobility in***
71 ***ombrotrophic peatlands***. Since the seminal paper of Shotyk et al. (1998), lead isotopic
72 signatures within accurately dated peat cores have shown to match not only the
73 signatures found in other archives (lake sediments, ice, cores, herbarium samples) but
74 also the isotopic signature of major Pb sources exploited/used at the time, i.e., isotopic
75 changes match periods of source or activity changes. In addition, because the isotopic
76 composition of Pb is a fingerprint of the source, Pb isotopes are a powerful tool to
77 distinguish natural from anthropogenic Pb sources (e.g. Shotyk et al., 1998; Mighall et
78 al., 2002; Baron et al., 2005; Shotyk and Le Roux, 2005; Kylander et al., 2006; De
79 Vleeschouwer et al., 2009a; Kylander et al., 2010; Martínez-Cortizas et al., 2016). If Pb
80 was mobile, Pb isotopes would not accurately reflect temporal variability in source
81 signatures and Pb isotopic changes in ombrotrophic peat cores would not correspond to
82 historical changes in metallurgical activities. Mobility would largely lead to isotopic
83 mixing, rendering it impossible to undertake reconstructions of Pb sources. However, the
84 above-mentioned studies showed that this is not the case.

85 D. **Careful chronological control of recent peat layers provides the evidence for Pb**
86 **immobility.** The distribution of ^{210}Pb (half-life: 22.26 years) in the uppermost peat layers
87 is indeed an argument for the preservation of Pb atmospheric signals in ombrotrophic
88 peat cores. Moreover, a multimethod approach allows a cross-check of ^{210}Pb age depth
89 models, and in most of the ombrotrophic peat cores the ^{210}Pb -based constant rate of
90 supply (CRS) age-depth models (Appleby and Oldfield, 1978; Appleby, 2001) are in
91 good agreement with pollen chronological markers (Appleby et al., 1997) as well as with
92 ^{14}C Bomb Pulse Curve, ^{137}Cs and ^{241}Am markers, and tephrochronological dating in
93 various locations (Goodsite et al., 2001; Givelet et al., 2003, Piotrowska et al. 2009; Li et
94 al., 2017; Davies et al., 2018).

95 E. **Experimental studies strongly support Pb immobility.** In a transplant study from a
96 polluted to an unpolluted site and *vice versa*, Novak et al. (2011) showed that Pb and
97 other trace metals are immobile in ombrotrophic peat, and that peat is therefore a
98 reliable archive of temporal pollution changes within a wide pH range (2.5–5.8). This
99 result confirmed an earlier study by Vile et al. (1999) who experimentally loaded ^{210}Pb
100 dated-peat cores with Pb solutions and Pb particulates which again demonstrated the Pb
101 immobility. When studying both solid and aqueous phases in three Finnish ombrotrophic
102 peatlands, Shotyk et al. (2016b) also demonstrated that Pb concentrations in the
103 aqueous phase never reached more than 0.01% of the total Pb at any given depth so
104 that the potential for post-depositional lead dissolution and mobility remains very small.
105 Moreover, these authors argue that the very limited vertical water movement in bogs
106 together with the size of the metal-containing particles in solution may also limit Pb
107 mobilization. They also found that the chronology of anthropogenic Pb isotopic
108 composition was similar in all three ombrotrophic peatlands over the past 1000yrs. Very
109 limited/negligible vertical Pb mobility in ombrotrophic peatlands has also been
110 demonstrated by down-washing experiments (Hansson et al. 2014, 2015b). This limited
111 mobility may have a small impact on calculated deposition rates for recent years but not
112 on decadal scales. In other words, such limited surface mobility may affect the

113 calculation of spatial Pb distribution, but not the use of Pb to reconstruct pollution history
114 over millennia.

115 F. ***By comparison to Pb, other trace metals such as zinc (Zn) are known to be mobile***
116 ***in many contexts and subject to post-depositional mobility or plant uptake***
117 (Twardowska et al., 1999; Rausch 2005a,b; Broadlay et al., 2007; Weiss et al., 2007).
118 Shotyk (1988) explained that Zn distribution is linked, among other factors, to
119 bioaccumulation, water-table fluctuations and pH conditions. Nieminen et al. (2002)
120 concluded that Zn is highly mobile in ombrotrophic peat, in contrast to well-preserved Pb
121 and copper (Cu) profiles, as Zn enrichment in peat was lower than that of Cu despite the
122 fact that Zn emission records from nearby smelters were significantly higher than those
123 of Cu. This corroborates other studies of Zn mobility in presence of organic matter
124 (Twardowska et al., 1999). Consequently, Kempter and Frenzel (2000) did not interpret
125 their Zn concentration profile in terms of anthropogenic record due to the possible
126 vertical migration of Zn. ***By contrast with Zn, the Pb profiles in ombrotrophic peat***
127 ***are often distinct from the typical vertical diffusion/advection and upward plant***
128 ***uptake profile of Zn*** (e.g. De Vleeschouwer et al., 2007, 2009a; Allan et al., 2013).

129
130 To conclude the evidence for Pb immobility, we once again urge Miszczak et al. (2019) to
131 carefully re-read the literature in the field before incorrectly re-interpreting other published
132 data. We also invite these authors to reconsider some of their unsupported assumptions. For
133 example: *“Binding strength of Pb, similarly as of other fugitive trace elements transported*
134 *with LRTP, in peat consisting predominantly of NOM (natural organic matter), at acidic pH is*
135 *not very high and this element is susceptible to release and vertical migration”* is an
136 unsubstantiated claim, especially considering that most of the Pb supplied to ombrotrophic
137 peatlands is in particulate form rather than as Pb^{2+} ions. If, however, Pb was supplied as Pb^{2+}
138 ions, it has previously been shown that *“soluble Pb^{2+} was retained in the peat through*
139 *physicochemical binding to organic matter, and as such Pb^{2+} was largely immobile in peat*
140 *even under conditions of a fluctuating water table”* (Vile et al., 1999). Hansson et al. (2015b)

141 also experimentally demonstrated the very limited Pb down-washing in ombrotrophic peat,
142 hence once again supporting the conclusion of functional immobility of Pb in ombrotrophic
143 peatlands.

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145 **3. TROPHIC STATUS OF PEATLANDS AND ELEMENTAL MOBILITY**

146 We would like to reiterate the importance of differentiating between ombrotrophic and
147 minerotrophic peatlands when reconstructing chronologies of past anthropogenic metal
148 pollution. Peatlands (or mires) can be divided in two end-member categories, defined by their
149 genesis, vegetation and hydrological functioning: ombrotrophic (also called “bogs”) and
150 minerotrophic (also called “fens”) peatlands. Of course, intermediate types exist, but there is
151 a tendency among peat geochemists to classify each site in one of these two categories.
152 Ombrotrophic peatlands are isolated from lateral inputs, and are therefore exclusively fed by
153 atmospheric inputs. In temperate areas such as Poland, their vegetation is dominated by
154 *Sphagnum* spp. together with lower amounts of other bryophytes, *Carex* spp. and
155 *Eriophorum vaginatum*. If undisturbed by drainage or cutting, water table fluctuation in those
156 sites is very low, and the pH is acidic (3-4). Minerotrophic peatlands also receive inputs from
157 the atmosphere but are additionally supplied by water from surrounding soils and rocks.
158 These lateral inputs, together with possible important water table fluctuations lead to an
159 important subsurface oxic zone and to a higher pH (from 5.5 to 8), conditions which are
160 favorable for metal mobility. The vegetation of such fens is characterized by the quasi-
161 absence of *Sphagna* species (which is indicative of ombrotrophy) and by the abundance of
162 other species such as *Equisetum* spp, *Carex* spp, *Menyanthes* spp. and *Molinia* spp.
163 (Charman, 2002). Unfortunately, Miszczak et al. (2019) seem to discount that fundamental
164 difference and therefore misuse past literature in minerotrophic or disturbed peatlands (Deiss
165 et al., 2004; Syrovetsnik et al., 2007; Smieja-Król et al., 2010, 2019) to argue for metal
166 mobility in ombrotrophic peatlands. Abundant literature based on studies in ombrotrophic
167 peatlands demonstrates their reliability in reconstructing chronologies of past atmospheric
168 metal pollution (Table 1).

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TABLE 1. Chronological order of selected literature data (n=78) reporting the immobility of lead in ombrotrophic peatlands (bogs) over the last 27 years.

Year	References
1993	Jones and Hao
1997	Brännvall et al., Farmer et al., West et al.
1998	Holynska et al., MacKenzie et al., Shotyk et al.
1999	Kempter and Frenzel, Martinez-Cortizas et al., Vile et al., Weiss et al.
2000	Kempter and Frenzel
2001	Shotyk et al.
2002	Martinez-Cortizas et al., Mighall et al., Nieminen et al., Shotyk, Shotyk et al
2003	Klaminder et al.
2004	Le Roux et al., Ukonomaanahoo et al., Bindler et al.
2005	Kylander et al., Le Roux et al., Shotyk et al., Shotyk and Le Roux
2006	Bindler and Klaminder, Bindler, Coggins et al., Kylander et al.
2007	De Vleeschouwer et al., Kylander et al., Weiss et al., Zacccone et al.
2008	Cloy et al.,
2009	Cloy et al., De Vleeschouwer et al., Kylander et al.
2010	Bao et al., De Vleeschouwer et al. a, Marx et al.
2011	Bindler; Bindler et al., Cloy et al., Klaminder et al., Novak et al., Silamikele et al., Zuna et al.
2012	Ferrat et al., Martinez-Cortizas et al., Meharg et al.
2013	Allan et al., Gallego et al., Pratte et al., Zinkutė et al.
2014	De Vleeschouwer et al., Fagel et al., Kuttner et al., Shuttelworth et al.
2015	Hansson et al., Shotyk et al., Veretennikova
2016	Martinez-Cortizas, Shotyk et al. a,b
2017	Hansson et al., Pratte et al., Talbot et al., Tudyka et al.
2018	Allan and Fagel, Borgulat et al., Fialkewicz-Kozziel et al., Perez-Rodriguez et al., Longman et al.
2019	Guo et al., Pawelczyk et al., Rosca et al., von Scheffer et al.

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4. **HOW TO USE LEAD DATA TO ACCURATELY RECONSTRUCT HISTORICAL CONTAMINATION**

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In this section, we emphasize best and widely accepted practices that guarantee accurate pollution reconstruction and outline the approaches to compare them to metal sources and emission records that were largely overlooked by Miszczak et al. (2019). We note the approaches we outline are not new, with these practices widely described within various papers, half of them being available via open access publications (e.g. Givelet et al., 2004; De Vleeschouwer et al., 2010b; Le Roux et al., 2010; Chambers et al., 2011).

186 **4.1. Sampling and subsampling – Data resolution and chronology**

187 The approach of Miszczak et al. (2019) compares their peat records to European Monitoring
188 and Evaluation Programme (EMEP) data, which are annual data on trace metal emissions
189 (<https://www.emep.int>). We detail here below why such approach requires a high-resolution
190 sub-sampling and chronological control.

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192 On a worldwide scale, Gorham (1991) suggested a value of 0.5 mm yr^{-1} as a conservative
193 estimate of the vertical accumulation rate of peat. For ombrotrophic peatlands, mean
194 accumulation rates have been estimated for various countries such as of Iceland (0.40
195 mm.yr^{-1}), Estonia (0.49 mm.yr^{-1}), Norway (0.18 mm.yr^{-1}), Finland (0.52 mm.yr^{-1}), Canada
196 ($0.48 - 0.50 \text{ mm.yr}^{-1}$) and Spain ($0.36-0.47 \text{ mm.yr}^{-1}$) (Everett 1983; Gorham, 1991; Mäkilä,
197 1997; Pontevedra-Pombal, et al., 2017). Peat accumulation rates as high as one centimeter
198 per decade have also been reported in a few European peatlands (Hughes et al., 2000;
199 Malmer and Wallén, 2004; Langdon and Barber, 2005; Pontevedra-Pombal et al., 2018).
200 Accumulation rates can vary depending on the vegetation type and climate conditions. For
201 example, cold periods may lead to a decrease in the peat accumulation rates (e.g. Charman
202 et al., 2013; Pontevedra-Pombal et al., 2018). As a result, high resolution sub-sampling and
203 dating is required if one would like to reconstruct decadal-scale atmospheric pollution.
204 However, Miszczak et al. (2019) present a low-resolution sampling and poor report on age
205 control, which precludes any detailed criticism and but raises a lot of doubts about the
206 chronological control of their data. Several papers have explained the advantages of high-
207 resolution sampling for trace metal reconstruction in peat (e.g. Givelet et al., 2004, De
208 Vleeschouwer et al., 2010b). The sampling strategy of Miszczak et al. (2019) does not meet
209 these widely accepted standards, with a sample resolution of 2.5cm at best. Part of the
210 problem is because they report a rather old sample set, from 1999. When the purpose is to
211 compare metal deposition rates in peat to high-resolution metal emission rates such as
212 EMEP, sample resolution is even more critical.

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214 Another major critical issue in the sampling approach adopted by Miszczak et al. (2019) is
215 the removal of surface vegetation. The authors have used the sampling protocol of Steinnes
216 et al. (2005). In that paper, it is said that “*Sphagnum moss ... and other plant material*
217 *growing on the surface were removed with a sharp knife before the coring, and the reference*
218 *surface level is thus the interface moss/peat. The thickness of the Sphagnum layer, if*
219 *present, was always less than 10 cm*”. This implies that these authors have removed the
220 living vegetation which is an integral part of the ombrotrophic peat deposit, and can
221 accumulate decades of information. This approach is poor practice as the critical importance
222 of keeping surface vegetation when reconstructing the chronologies of anthropogenic
223 pollution using ombrotrophic peat sections has been abundantly demonstrated (e.g. Givelet
224 et al., 2004; Farmer et al., 2006; Kempter et al. 2007; Le Roux et al, 2010; De Vleeschouwer
225 et al., 2010b). Removing the surface vegetation and taking the peat/vegetation interface as
226 year 1999 is incorrect and leads to large uncertainties on any chronology or inventory
227 calculations performed thereafter. As Givelet et al. (2004) pointed out: “*the historical record*
228 *of atmospheric Pb deposition is so well preserved in undisturbed bogs that our ability to read*
229 *and interpret the peat bog records is largely independent of chemical processes taking place*
230 *within the bog itself, but can depend to a large extent on the methods used to collect, handle,*
231 *and prepare the samples for analysis*”. We therefore question how Miszczak et al. (2019) can
232 accurately compare multi-centimeter samples that represent several tens of years with
233 EMEP data - itself only covering the past 40 years - while the peat samples were collected in
234 1999, barely 20 years after the inception of EMEP data (1980), and while up to 10cm of the
235 top vegetation were removed (possibly representing up to 20 years of information). This
236 method therefore removes the deposits that could have been compared to those 20 years of
237 EMEP acquisition.

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240 **4.2. Interpreting data in elemental ratios, enrichment factors (EFs) and metal**

241 **accumulation rates, certainly not in concentration**

242

243 Another major concern with the paper published by Miszczak et al. (2019) is their mis-
244 interpretation of other published data, in particular the data of De Vleeschouwer et al.
245 (2009a). The authors claim that De Vleeschouwer et al. (2009a) ignored the similarity
246 between Pb concentration and density profiles, a similarity that Miszczak et al. (2019) claim
247 to be evidence of Pb post-depositional mobility. Miszczak et al. (2019) seem misunderstand:
248 1) the record presented in De Vleeschouwer et al. (2009b) and why the density and
249 accumulation rates change along the peat profile of Slowinskie Blota, and 2) how to use Pb
250 as a tracer of past anthropogenic activity. We rebut their mis-interpretation in more detail
251 below.

252

253 **4.2.1. Density, accumulation rate and Pb concentration.**

254 Firstly, typically peat accumulation is interpreted chronologically, that is, from bottom to top,
255 and not the opposite as Miszczak et al. (2019) do. There is indeed a period of higher bulk
256 density in Slowinskie Blota, from 50 to 35 cm depth. This period corresponds to the Little Ice
257 Age as reported in De Vleeschouwer et al. (2009b). During this period, the colder
258 temperatures resulted in a decrease in plant growth and a subsequent decrease in the peat
259 accumulation rates. In addition, this period saw increased windiness and dune activity that
260 resulted in increased aeolian lithogenic input to this ombrotrophic peatland (De
261 Vleeschouwer et al. 2009b). This combination of reduced accumulation rates and increased
262 dust input resulted in an increase in the bulk density. The peat concentration profile is
263 therefore affected by this decrease in accumulation rates (i.e. concentration effect – apparent
264 Pb concentration increase due to accumulation rate decrease) in addition to displaying an
265 increase in the concentrations of pollutant elements from the industrial Revolution. Indeed,
266 the maximum Pb concentrations in the De Vleeschouwer et al. (2009a) dates from the 1960-
267 70s, coinciding with maximum Pb emissions, in particular from leaded gasoline, just prior to
268 its banning, which began in the 1980s (Schneider et al., 2000; Pacyna and Pacyna, 2000;

269 von Storch et al., 2003). The Pb peak has therefore nothing to do with post-depositional Pb
270 migration.

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272 **4.2.2. Pb as a tracer of past anthropogenic activity**

273 Pertinently, De Vleeschouwer et al. (2009a) do not use Pb concentrations to interpret periods
274 of past anthropogenic activities. Since the pioneering papers of Lee and Tallis (1973) and
275 Livett et al. (1979) who reported Pb pollution trends, practices have changed, both showing
276 that Pb was immobile, but also in finding accurate ways of using trace metal data to
277 reconstruct past environmental pollution. It is irrelevant to use raw concentrations to
278 reconstruct past anthropogenic activities. The reason is simple and already mentioned
279 above. The matrix effect of the peat itself alters apparent Pb concentrations: i.e. if the
280 accumulation rate increases, it will dilute the concentration of a given element in sample and
281 *vice versa*. To avoid this dilution effect, it is common practice to use metal to lithogenic
282 element ratios, i.e. Enrichment Factors (EFs), or metal accumulation rates (e.g. Shotyk et al.,
283 1998; Le Roux et al., 2010; Allan et al., 2013; De Vleeschouwer et al., 2014). Additionally,
284 not all the Pb deposited in a peatland represents pollution, as there is always a component of
285 Pb supplied from dust deposition. Using EFs also allows the Pb from anthropogenic
286 contamination to be separated from natural dust.

287

288 Again, Miszczak et al. (2019) seem to overlook these important considerations and mis-infer
289 the data from De Vleeschouwer et al. (2009a) who critically based their interpretation on Pb
290 EFs and accumulation rates (see figure 4 of De Vleeschouwer et al., 2009a). By accurately
291 using Pb accumulation rates, De Vleeschouwer et al. (2009a) successfully reconstruct the
292 history of anthropogenic lead in Poland. This traces Medieval lead mining, followed by the
293 apogee of the Polish Renaissance, periods of wars and trouble that see the decrease of
294 mining activities, culminating with the boom of the Industrial Revolution and the ban of Pb-
295 gasoline. Using this accurate approach (high-resolution sampling and dating including
296 surface vegetation, use of EFs and Pb accumulation rates), De Vleeschouwer et al. (2009a)

297 demonstrated that the Pb accumulation rate obtained in “..the top-most (i.e. surface
298 vegetation) sample ($1 \text{ mg m}^{-2} \text{ yr}^{-1}$) was in very good agreement with the lead deposition
299 scenario (between 1.1 and $1.2 \text{ mg m}^{-2} \text{ yr}^{-1}$) provided by the Meteorological Synthesizing
300 Centre-East (www.msceast.org) and with lead wet deposition ($1.16 \text{ mg m}^{-2} \text{ yr}^{-1}$) in this region
301 of Poland measured by EMEP (www.emep.int) in 2005 (the year when the core was
302 collected).” The record of De Vleeschouwer et al (2009a) is backed up with Pb isotopes that
303 confirm the history of anthropogenic Pb by identifying the sources of Pb in Poland
304 (metallurgy, coal, gasoline), and is yet another record amongst many others (Table 1) that
305 demonstrates Pb to be immobile in ombrotrophic peatlands and an excellent a recorder of
306 anthropogenic activities when properly used. This last point is again amongst best practice:
307 the robustness of Pb pollution reconstructions and Pb immobility in ombrotrophic peatlands is
308 strengthened when based on an array of geochemical (Pb accumulation rates, EFs on
309 several ratios, Pb isotopes) and further evidences (micro and macrofossils,
310 historical/archaeological data on mining, etc.) instead of one Pb concentration profile.

311
312 To finalize our concerns, we would like – on the top of the points raised above - to draw the
313 reader’s attention on a series of questionable assumptions in Miszczak et al. (2019). For
314 example, “the uppermost 0–5/7.5 cm layer of peat cores was mostly formed over the last
315 150–200 years” or “The 20–40 cm peat layer usually represents an age of 500–1500 years
316 when there were no extensive sources of anthropogenic air pollution” or “the deeper peat
317 layers in the studied bogs represent an age over 300 up to 2000–3500 years BP, when there
318 were no extensive sources of anthropogenic air pollution”. These assumptions are not
319 supported by the data. Raw radiocarbon dates are not reported, making it impossible to
320 check these assumptions. Furthermore, for Polish sites, Miszczak et al. (2019) do not have
321 any chronological control over the last 150 years such as ^{210}Pb , ^{137}Cs and ^{241}Am dating. This
322 makes such assertions highly questionable. Moreover, the assumption that there is no
323 anthropogenic activity before 300 years BP is largely erroneous, as evidenced by a multitude
324 of other records in Poland (De Vleeschouwer et al., 2009a; Tudyka et al., 2017, Fialkewicz et

325 al, 2018) and across Europe (e.g. Shotyk et al., 1998, Kylander et al., 2005; Le Roux et al.,
326 2006; Martinez-Cortizas et al., 2002, 2016; De Vleeschouwer et al., 2010a; Longman et al.,
327 2018) as well as long distance ice core evidence such as in Greenland (e.g. Rosman et al.,
328 1997). Similarly, we question the unsupported assumption that “*The LRTP deposition*
329 *resulting from anthropogenic activities, should have been concentrated in the uppermost*
330 *layers of peat cores that were formed since 1780/1848, i.e. over the last 150–200 years BP.*”
331 On what is such an assumption based? It is unfortunately impossible to know. What we do
332 know, however is that Medieval atmospheric pollution was great across in Europe and on
333 occasion greater than during the Industrial Revolution (Kempter and Frenzel, 1999).

334

335

336 **5. CONCLUSION**

337 A combination of what we see as erroneous and unsupported assumptions drawn from an
338 old dataset together with a mis-interpretation of literature data provided the impetus for us to
339 write this comment on Miszczak et al. (2019). It is important that future readers and young
340 scientists continue to document trace metal reconstruction of anthropogenic activities in
341 environmental archives using appropriate and accurate techniques. Our review contrasts the
342 claim of Miszczak et al. (2019) that Pb deposition chronologies cannot be reconstructed due
343 to bulk density-affected element vertical redistribution. We moreover hope that this comment
344 brings back into focus the last 40 years of literature in the field largely overlooked by
345 Miszczak et al. (2019). This body of work reiterates time and again that Pb is immobile in
346 ombrotrophic peatlands (or bogs) and that it is a reliable tool for reconstructing past natural
347 changes in Pb as well as anthropogenic activities. These numerous studies make use of
348 multiple tools such as metal to lithogenic element ratios, enrichment factors and/or metal
349 accumulation rates combined to high-resolution sampling and chronology. Coupled with Pb
350 isotopes, it is possible to decipher emission sources at a regional to continental level. The
351 numerous papers in the field are supported by similar reconstructions in other environmental
352 archives (herbarium samples, lake sediments, ice cores) as well as experimental

353 approaches. These reconstructed anthropogenic activities, thanks to the immobility of Pb in
354 ombrotrophic peatlands, are unequivocal indicators of the Pb contamination of our planet and
355 we anticipate many more papers to come on Pb and other trace metal contamination in the
356 environment.

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