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**Nd isotopes as a tracer of dust trapped by peat bogs: A reminder of basics. Comment on: « Neodymium isotopes in peat reveal past local environmental disturbances » by Marcisz et al. (2023)**

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Neodymium (Nd) isotopes are one of a suite of radiogenic isotopes whose ratios in any geomaterial constitute a sensitive and faithful fingerprint signature of the material's original reservoir. A plethora of studies have demonstrated the relevance of Nd isotopes as powerful geochemical tracers of rock and sediment provenance at various geological time scales (e.g. O'Nions et al., 1983; Goldstein et al., 1984; Grousset et al., 1988; Dia et al. 1992). More recently, they have been applied to a range of palaeo-source studies, including palaeoceanography (e.g. Bazhenova et al., 2017; Fagel et al., 2004; Frank, 2002; Grousset et al., 1992; Lacan and Jeandel, 2004) and palaeo-dust studies (e.g. Aarons et al., 2016; Beaudon et al., 2022; Grousset et al., 1992; Longman et al., 2022; Saitoh et al., 2011; Sharifi et al., 2018; Simonsen et al., 2019; Stewart et al., 2023; Struve et al., 2022). However, since the first Nd isotope study by Le Roux et al. (2012), only a few studies have been published in peat bogs (e.g. Allan et al., 2013; Fagel et al., 2014; Fialkewicz-Koziel 2016, Pratte et al., 2017, Sharifi et al., 2018; Vanneste et al., 2015, 2016). There are several reasons for this: firstly, Nd isotopes must be measured using multicollector mass spectrometer such as MC-ICP-MS (Multicollector Inductively Coupled Plasma Mass Spectrometer) or TIMS (Thermal Ionization mass Spectrometer), which are not readily available in every laboratory. Secondly, Nd has to be separated by a chemical process to ensure that there are no Rare Earth Elements (REE) interferences. These procedures are not available to everyone, expensive and time consuming. Another simpler reason is that the dust trapped by peat bogs can be traced by other, more easily measured proxies, and that in fact the use of Nd isotopes is only the final step in a long process of geochemical characterization of the past atmospheric fallout in a peat bog. These other tracers are: lithogenic and REE concentrations and ratios, acid insoluble ash content or mineral characterization by XRD, SEM or grain size analyses. Presently, artificial intelligence has also been employed (see our recent dialog with Chat GPT in S.I.).

Prior to any inorganic geochemical investigation in peat aiming at reconstructing atmospheric dust fluxes (mineral dust or pollutants), one should apply rigorous site selection criteria. This is because atmospheric deposition reconstruction using peat bogs is based on 2 paradigms:

1. The selected site is ombrotrophic (i.e. it is a peat bog, not a minerotrophic fen): it is exclusively fed by atmospheric inputs, in very acidic (pH generally below 4) and anoxic conditions;
2. The dust that is deposited in a peat bog does not - most of the time - represent local dust, but instead originate from various distal sources.

The first paradigm has been verified many times by independent geochemical comparisons, in particular the comparison between airborne pollutants measured in precipitation and herbaria vs. records in peat bogs or mosses (e.g. Bindler, 2011; Dunlap et al., 1999; Farmer et al., 2002; Kempter et al., 2010, 2017; Markert, 1991; Novak et al., 2008; Le Roux et al., 2005; Pech et al., 2022; Rausch et al., 2005; Shotyk et al., 2015; Steinnes, 1995; Weiss et al., 1999, 2002; Zupančič and Bozau, 2022). It should be noted that simply the presence of *Sphagnum* moss occurrence in a peatland does not systematically indicate the site is ombrotrophic. Important differences can occur between the botanical and geochemical trophic status of a peatland because proxies are forming/accumulating in different ways; this was recognized several decades ago (e.g. Shotyk et al., 1998, 2002). Therefore, in any atmospheric reconstruction study using a peat bog, the geochemical trophic status of a site must always be assessed beforehand using an array of geochemical indicators (e.g. major and trace elements abundances and ratios, mineralogy, pH etc.) and a topographic assessment to exclude the possibility of local mineral inputs prior to embarking on a reconstruction of atmospheric sources of dust (e.g. De Vleeschouwer et al., 2007, Fialkewicz-Koziel, 2018, 2020, 2022; Li et al., 2020; Longman et al., 2017; Pratte et al., 2016, 2017a,b, 2020). All other types of peatland records (i.e. not ombrotrophic) should be considered as 'classical' sediment archives. As such, any atmospheric input will be mixed or possibly overprinted by lateral mineral inputs from the catchment, elemental diffusion from the bedrock, etc. 'Classical' sediment archives make it extremely challenging (virtually impossible) to separate an atmospheric signal from local inputs. Records from minerotrophic peatlands should be considered with caution and supported by additional geochemical proxies such as loss-on-ignition (i.e. ash weight-%), lithogenic element (Al, Ti, Sc, REE) concentrations, mineral identification and/or Principal Component Analysis (PCA), to support an atmospheric proxy reconstruction (e.g. De Vleeschouwer et al., 2014; Vanneste et al., 2015; Fagel et al., 2014; Fialkewicz-Koziel et al., 2022; Sharifi et al., 2018). However, Marcisz et al. (2023) have ignored these key points, and based their study on a kettle hole peatland, which could be geochemically minerotrophic (e.g. Lindsay et al., 2015) as well as having top-down peat accumulation from vegetation rafts (Gaudig et al., 2006). Their site is furthermore disturbed by several fires and floods (that can provide lateral mineral inputs disturbing a possible atmospheric signal) as mentioned by the authors. Marcisz et al. (2023) then make a rather speculative conclusion from a single atmospheric dust proxy without providing any geochemical evidence of ombrotrophy. The only proxy presented by Marcisz et al. (2023) that would help the reader assess the geochemical trophic status of the peatland, although not sufficient because not geochemical, is the ash content, but the authors incorrectly report the data in grams instead of in weight-%. This makes it impossible to evaluate the organic content (and therefore the mineral content) of the core. Loss of organic content reported in grams is strongly affected by variations in the peat accumulation rate. Ignoring this basic concept and reporting an ash content in grams removes any interpretative value from the data. One can however estimate the LOI percentage in Marcisz et al. (2023) peat core from the ash weight and ash free density reported in figure 3 (and knowing that 2cm<sup>3</sup> were taken for these measurements). This would lead some levels to have an ash/mineral content of 75% which question if those levels are peat (see SI for detailed calculation).

The second paradigm, first expressed by Mattson and Koutler-Anderson (1954), is that peat bogs are located in humid climate zones, where a cover of native vegetation stabilizes the soil and, therefore, minimizing wind erosion and surface runoff. For example, Aubert et al. (2006) have shown that despite the pronounced geological differences between two peatlands located in the Black Forest (granite bedrock) and the Jura (limestone bedrock), similar REE signatures and deposition rates were found in both peatlands. Given that both sites confirm an Upper Continental Crust signature, the data suggests both sites are influenced by regional and not local, soil-derived lithogenic aerosols. In the case of the Etang de la Gruère study (Le Roux et al., 2012), the term local corresponds to the catchment area (10 km), regional to 100 km and global to thousands of km (as defined by Lawrence and Neff, 2009). The larger the peatland and the more it is in a protected area, the less risk there is for recent inputs of local dust. For example in his seminal studies, Gorham foresaw this, based on a transect of a bog to a road and a transect of collected *Sphagnum* mosses in bogs in Minnesota, where high concentrations of lithogenic elements were associated with the presence or absence of cultivated land (Gorham and Tilton, 1978; Santelmann and Gorham, 1988). It should also be noted that local sources have sometimes

been identified in dust records from peat bogs (e.g. De Vleeschouwer et al., 2009; Vanneste et al. 2015) but those local sources had been identified beforehand by studying the geographical and geomorphological settings, sometimes supported by geochemical analyses of the local source itself to make sure to decipher it from potential distal dust sources (e.g. Vanneste et al., 2015, 2016).

By measuring Nd isotopes alone (i.e. with no other geochemical proxies as mentioned above) in a peatland without an in-depth geochemical study of the trophic status (i.e. possibly geochemically minerotrophic), Marcisz et al. (2023) ignore two widely accepted paradigms and do not use the standard practice supported by a large body of literature and approved by the community of peat geochemists. Without a proper geochemical characterization of their record (e.g. lithogenic elements, insoluble ash content, mineralogy, grain size, PCA to determine the driver of each element's behavior) as well as the geomorphological and geographical setting of their site (i.e. lateral/local mineral inputs), it is impossible to assess the suitability of their record for atmospheric reconstruction. In addition, any mixing process of dust sources could be identified if the Nd isotopic data were supported by elemental composition/ratios and by other isotopic systems (for example Pb isotopes). But Marcisz et al. (2023) have employed Nd as a stand-alone proxy, therefore, any interpretation not supported by any in-depth geochemical characterization of the records is speculative.

Given that, we are therefore rather surprised by the unsupported criticisms of Marcisz et al. (2023) towards our previous studies in paragraphs 4.2 and 4.3, for which we provide a point-by-point rebuttal in the supplementary information. We would however like to highlight two facts that are valid for our studies:

- Long-term peat records do indeed provide local signatures by having access to the deeper sediments (whether it is minerotrophic peat or lake sediments), thus providing a geochemical and isotopic signature of the catchment. This is the case for example for Le Roux et al., (2012) and Stewart et al (2023) where the basal sediment has a clear distinct signature from upper peat layers.
- Our studies always provide complete peat records, i.e. it includes the upper part of the profile characterized by living or poorly decomposed moss layers

In addition, we would like to point out that Nd isotopes have been successfully used as cryptotephra identifiers in various archives and time scales (e.g. D'Antonio et al., 2016; Hart et al., 1993). Moreover, the distinction of young volcanism from old shields is somewhat basic for the use of Nd isotopes as provenance tracers. The Nd isotopic signature of a rock is indeed primarily controlled by its age, with fresh mantle-derived rocks having a much more positive  $\epsilon_{Nd}$  than older cratonic rock (Bizimis & Scher, 2016). Therefore, the isotopic compositions of Nd (as well as Sr and Pb) radiogenic isotopes are significantly different in mantle- vs. in crust-derived rocks, permitting the distinction between the two extreme origins and of lithologies in between: young volcanic areas and old continental shields (Grousset and Biscaye, 2005). Marcisz's et al. (2023) seem however to somehow ignore these critical basics of radiogenic isotopic geochemistry. Moreover, Nd isotopes have also been used elsewhere to distinguish two distal dust sources with different origin (volcanic Iceland dust vs Saharan dust) in peat deposits (Stewart et al., 2023). In the case of the Vasset-Killian tephra, it has moreover been previously identified in Swiss lakes and French peatlands (e.g. Hajdas et al., 1993, Hajdas and Michczynski, 2010, Juvigné, 1991). The unambiguous volcanic  $\epsilon_{Nd}$  and REE signature reported by Le Roux et al. (2012) is supported by an array of evidence: literature data showing that eruptive rocks from the "Chaîne des Puys" (where the Vasset-Killian complex belongs) display positive Nd signatures (e.g. Hamelin et al., 2009), Nd isotopes, peak in ash content and density, and ultimately radiocarbon dating.

In conclusion, Nd continues to be a very useful and sensitive geochemical tracer in peat bogs, but it is dependent on choosing a suitable site where one can have a high degree of confidence that any mineral material separated from the peat should be from dust sources transported atmospherically. This should be assessed by a range of geochemical and mineralogical proxies to characterize the studied site to avoid any erroneous interpretations. Finally, reference material from potential local and distal dust sources must be isotopically characterized and their pathways to the site either excluded (as for local sources) or atmospheric routes understood (as for distal sources).



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## Supplementary information

### S.I.1 : Chat GPT 05/02/2023

#### *How to investigate dust in peat paleoenvironmental records?*

There are several methods to determine if dust in peat paleoenvironmental records is of local origin or from a remote source:

**Elemental and isotopic analysis:** Comparing the elemental and isotopic composition of the dust to local soils and sediment, as well as known remote sources, can help identify its origin. For example, if the dust contains elements that are not typically found in the local area, but are common in known remote sources, it is likely that the dust was transported from a remote location.

**Statistical analysis:** Using statistical methods such as principal component analysis (PCA) or clustering algorithms, can help identify patterns and trends in the elemental and isotopic composition of the dust, which can help distinguish between local and remote sources.

**Physical analysis:** Examining the size and shape of the dust particles can help determine if they were transported over long distances. Dust particles that are larger and more angular are typically indicative of local sources, while smaller and more rounded particles are often associated with remote sources.

**Comparison with other paleoclimate proxies:** Comparing the results of the dust analysis with other paleoclimate proxies, such as ice cores, tree rings, and sediment records, can help confirm the origin of the dust. For example, if the dust is found in association with other proxies that are known to be from a remote source, it is likely that the dust is also from that source.

**Modeling:** Using atmospheric circulation models can help simulate the transport of dust from remote sources and predict the chemical and isotopic composition of the dust that would be expected to arrive at a particular location. This information can be compared to the actual elemental and isotopic composition of the dust in the peat record to help determine its origin.

#### *Can we use Nd isotopes in peat to evaluate dust sources?*

Yes, Nd (neodymium) isotopes can be used to evaluate dust sources in peat paleoenvironmental records. Nd isotopes are a valuable tool for tracing the sources of sediment in the environment because different geological regions have unique Nd isotopic signatures. By analyzing the Nd isotopes in the dust, it is possible to determine the source of the dust and whether it is from a local or remote source.

In peat records, Nd isotopes can be measured using techniques such as inductively coupled plasma mass spectrometry (ICP-MS). The Nd isotopic composition of the dust is then compared to the Nd isotopic composition of local soils and sediment, as well as known remote sources. A match with a known remote source would suggest that the dust was transported from that location, while a match with local soils and sediment would suggest a local origin.

**It is important to note that while Nd isotopes can be a useful tool for evaluating dust sources, they should be used in conjunction with other methods, such as elemental analysis and physical analysis, to provide a more complete picture of the origin and transport of the dust.**

### **S.I.2: attempt at calculating an ash content (in weight %)**

In figure 3 of their paper, Marcisz et al. (2023) report various parameters that are not the common practice:

- “ash free density” instead of bulk density. Our understanding is that this density was derived from the sample after loss on ignition was performed. In other words that the mineral content of the sample was “removed” to calculate a density that only represent the organic matter content.
- Ash content is reported in grammes instead of weight %, which renders impossible to estimate the real proportion of mineral in the peat.

Marcisz et al. (2023) however report that 2cm<sup>3</sup> were taken to perform LOI measurements. Reading from figure 3, there are levels containing 0.9g of ash with an ash free density of 0.15g cm<sup>-3</sup>. That means a 2cm<sup>3</sup> sample is composed of 0.3g of organic matter and 0.9g of ash (i.e. ash/mineral content is 75%), which is not peat. It is possible we misunderstand the proxies reported by Marcisz et al. (2023), but this simple calculation is not made to prove that the studied core is not only composed of peat, rather than to show that once standard practices are not used, the reader is forced to navigate to try calculate a very basic proxy, while it would have just been very simple to report bulk density and ash content in weight-% to allow the reader to have a proxy of the geochemical trophic status of the peatland.

### **S.I.3: Point by point answer to paragraphs 4.2 and paragraphs 4.3 of Marcisz et al. (2023):**

We agree that there seems to be a misquotation on the Allan et al. (2013) paper but as the main author is no longer in academic research, we do not wish to comment on that paper without his consent. We will therefore focus on the Etang de la Gruère record (Le Roux et al., 2012).

*« However, these studies did not include reference samples collected in close proximity to the peatlands, and they refer solely to  $\epsilon$ Nd data from the literature, measured at locations distant from the investigated sites (Allan et al., 2013; Fagel et al., 2014; Le Roux et al., 2012). »*

As stated in the main text, In the 3 cited papers, we were able to collect the deepest sediments representing the geochemical signature thus the « local » signature, or that is to say, the average signature of the close proximity of the peatland.

*« Moreover, the authors did not define what they mean by a “local source”, and how far from the site this local source may be located. For example, Le Roux et al. (2012) attributed the observed variability in the  $\epsilon$ Nd values to shifts between a local source and Saharan dust, but, in the absence of additional data, provided no unequivocal evidence for the contribution of the latter. »*

We deplore the cherry-picking of this paragraph by Marcisz et al. (2023), where the authors ignore decades of publications on Etang de la Gruère where lithogenic elements and Pb isotopes also show dust provenance from Sahara (see for example: Shotyk et al., 1998; 2002). In addition, it is well known that Sahara dust is a major dust source to European mountains (Bacardit and Camarero, 2009; De Angelis and Gaudichet, 1991) including the Alps, where Etang de la Gruyère is located.

*« A comparison of these records shows that it is practically impossible to identify Saharan dust supply to a peatland based only on Nd isotopes alone; other, supporting evidence is needed to unequivocally detect Saharan derived materials. »*

We fully agree that Nd alone is not enough (we note that it is however precisely what Marcisz et al. (2023) do: they only measure Nd isotopes, and therefore their argument is circular). That is why all our studies are based on a range of proxies before measuring Nd isotopes, such as: inorganic elemental geochemistry including metals and REE, grain size/mineralogy, acid insoluble ash content, Pb isotopes, etc.

*« The apparent coincidence of a change in the  $\epsilon\text{Nd}$  values and a reconstructed age of a specific peat layer is not direct evidence, as age-depth models are often not precise enough to allow such direct comparisons »*

Once again Marcisz et al. (2023) were cherry-picking, and did not take the time to our paper in detail, especially the SI which details the work on age-depth modelling, including the comparison of the two Etang de la Gruère cores. In particular, the age-depth model developed for the master core shows sufficient resolution to detect centennial-scale events.

*« Le Roux et al. (2012) attributed their Nd isotope record ( $\epsilon\text{Nd}$  values of  $-9.7$  and  $-9.6$ ) to a supply of the Icelandic Vedde Ash, but this interpretation was supported only by the age of the peat layer in which the change in the  $\epsilon\text{Nd}$  values occurred. »*

This is again a misreading of our paper by Marcisz et al. (2023). We wrote: « The second peak (ca. 12 kyr B.P.) is a positive shift that **may** correspond to a volcanic signature (Vedde Ash) ». Moreover, not only do we have the Nd isotopic evidence, but this layer also has a density, ash content, and dust flux peak as well as distinct REE signatures suggesting a volcanic origin. This layer has already been attributed to a volcanic eruption by Shotyk et al (1998, 2002).

*« Similarly, the highest measured  $\epsilon\text{Nd}$  value of  $-1.0$  was assigned to two eruptions from the Massif Central, because of the similar age of this peat layer, rather than the similarity of the  $\epsilon\text{Nd}$  signatures (Le Roux et al., 2012). »*

Here Marcisz et al. (2013) questions a tephra layer which is well established in the literature and obviously did not take the time to indeed check the literature we provide in our paper. This tephra was for example identified in peat bogs from Massif Central (Juvigné, 1991) and in sediments from Lake Soppensee (e.g. Hajdas et al., 1993, Hajdas and Michczynski, 2010). The Nd isotopic signature is without doubt volcanic (e.g. Grousset and Biscaye, 2005). Moreover, the Killian-Vasset Complex is part of the “Chaine des Puys” which eruptive rocks display positive  $\epsilon\text{Nd}$  signatures up to +3.8 (eg. Hamelin et al., 2009) and there is no volcanic terrain in the peatland area since the watershed’s bedrock is mainly composed of carbonates (Shotyk et al., 1998).

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