Norsk institutt for luftforskning
Norwegian Institute for Air Research

Review of the Assessment of Industrial Emissions with Mosses

Stephen Michael Mudge, Katrine Aspmo Pfaffhuber and Hilde Thelle Uggerud

NILU report 22/2019

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<tr><td>NILU report 22/2019</td><td>ISBN: 978-82-425-2991-6
ISSN: 2464-3327</td><td>CLASSIFICATION:

A – Unclassified (open report)</td></tr>
<tr><td>DATE

01.10.2019</td><td>SIGNATURE OF RESPONSIBLE PERSON

Ole-Anders Braathen, Deputy Director (sign.)</td><td>NUMBER OF PAGES

26</td></tr>
<tr><td colspan="2" rowspan="2">TITLE

Review of the Assessment of Industrial Emissions with Mosses</td><td>PROJECT LEADER

Hilde Thelle Uggerud</td></tr>
<tr><td>NILU PROJECT NO.

O-119086</td></tr>
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<tr><td colspan="3">ABSTRACT
Commissioned by Norwegian Environmental Agency, NILU - Norwegian Institute for Air Research has surveyed the literature on the topic of “Assessment of industrial emissions using moss”. The purpose is to provide an overview of published knowledge on possible relationships between metal concentrations in moss and air quality, emissions, uptake in other organisms and impacts on environment and health. In addition, there was a request for information on whether other countries use moss surveys around industries and, if so, how the results are used by the authorities. The literature search resulted in 51 relevant publications, which mostly are from the period 2016-2019. The results of these publications show that moss is a good passive sampler for airborne contaminants and can provide valuable information on chemical signature and deposition of metals. No studies have been found that relates concentration in moss to air quality or amount emission from selected industries. A single 2019 study attempts to link moss concentration in context of health effects. A survey among the participating countries in ICP-Vegetation shows that results from moss surveys so far not have been used by authorities in a regulatory context.</td></tr>
<tr><td colspan="3">NORWEGIAN TITLE

Litteraturstudie om vurdering av industriutslipp ved bruk av mose</td></tr>
<tr><td colspan="3">KEYWORDS</td></tr>
<tr><td>Metals</td><td>Moss</td><td>Emissions</td></tr>
<tr><td colspan="3">ABSTRACT (in Norwegian)
På oppdrag fra Miljødirektoratet har NILU - Norsk institutt for luftforskning, gjort en litteraturstudie innenfor temaet «Vurdering av industriutslipp ved bruk av mose». Hensikten er å framskaffe en oversikt over hva som er publisert av kunnskap om eventuelle sammenhenger mellom metallkonsentrasjoner målt i mose og utslippsmengder, luftkvalitet, opptak i andre organismer og betydning for miljø og helse. Det er i tillegg etterspurt informasjon om hvorvidt andre land benytter moseundersøkelse rundt industri og eventuelt hvordan disse resultatene blir brukt av myndigheter. Litteratursøket resulterte i 51 relevante publikasjoner hvor de fleste er fra perioden 2016-2019. Resultatene fra disse publikasjonene viser at mose er en god passiv prøvetaket for luftforurensinger og kan gi verdifull informasjon om kjemisk signatur og deposisjon av metaller. Det er ikke funnet noen studier som relaterer konsentrasjon i mose med luftkvalitet eller mengde utslipp fra utvalgte industrier. En enkelt studie forsøker å sette mosekonsentrasjoner i sammenheng med helseeffekter. En spørreundersøkelse blant deltakerland i ICP-Vegetation viser at resultater fra moseundersøkelser så langt ikke er benyttet av myndigheter i reguleringssammenheng eller lovgivning.</td></tr>
<tr><td colspan="2">PUBLICATION TYPE: Digital document (pdf)</td><td>COVER PICTURE: Source: NILU</td></tr>
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Contents

1 Background

There have been eight national moss surveys to study the atmospheric deposition of metals in Norway since 1977 (*e.g.* Steinnes *et al*., [1-3]). In 2000, the first additional survey around selected industries was conducted to assess metal deposition at the local scale near potential sources. Reports [4-7] from these industry surveys reported concentrations, but lack information regarding the connection between concentration levels measured in moss and emissions, air quality, uptake in other organisms and whether the concentration levels in moss is reason for concern. This current report is a synthesis of published literature and conversations with European colleagues working with moss biomonitoring assessing whether these relationships have been established in other studies.

2 Approach

2.1 Literature search

Keywords including moss and accumulation / industry *etc*. were used in a Web of Science search and 50+ peer-reviewed scientific papers were downloaded as PDF-files for review. The papers presented results from 20+ countries mostly from the past four years (2016 – 2019). The focus was on how moss samples were used to assess industrial emissions. The results are summarised according to the different sections below that provide answers to the questions posed. Key references and their purpose can be seen in the Appendix.

It should be noted that the phrase "chemical signature" is frequently used in describing the composition of elements in mosses. The signature is independent of the concentration and is the internal relationship between the elements measured. Two mosses may have vastly different concentrations, but the signatures may be the same and conversely, two mosses may have similar concentrations, but the internal relationship between the metals may be completely different indicating a different source. In reality, almost all chemical signatures identified in mosses will be a mixture of background / baseline conditions with one or more contaminating additions on top. Multivariate statistical methods will be required to differentiate between the different contributors and apportion the proportion from each source in the moss.

2.2 Survey among countries participating in ICP-Vegetation

Information on how authorities in other countries use data from moss surveys around industries is not necessarily found among scientific publications. In order to obtain this type of information, the ICP-Vegetation network was used. ICP-Vegetation is an international research programme investigating the impacts of air pollutants on crops and (semi-)natural vegetation. Part of the programme focuses on the atmospheric deposition of heavy metals, nitrogen and persistent organic pollutants (POPs) to naturally growing mosses. ICP-Vegetation reports to the Working Group on Effects (WGE) of the UNECE Convention on Long-Range Trans-boundary Air Pollution (LRTAP). The aim of the European moss survey is to identify areas with enhanced concentrations of heavy metals and the reported data are used in assessment of the current environmental state and to predict the expected future environmental state.

A request was sent to the ICP-Vegetation chairman (United Kingdom) to find out if moss data are used in any legislation within UNECE. In addition, the request was sent to key participants (excl. Norway) in the moss society, which are Albania, Austria, Czech Republic, Estonia, Finland, France, Germany, Iceland, Italy, Latvia, Lithuania, Macedonia, Poland, Romania, Russia, Serbia, Slovenia, Sweden and Switzerland to obtain information on a national level. Responses were received from 10 out of 20 participants (incl. chairman). The questions asked were:

1. How do authorities in your country use the data from the moss surveys around industries?
2. Are the data used in any form of legislation?

The results to these questions are summarised below.

3 How are mosses currently used?

Mosses are used as passive samplers of particles since they have no root system that acquires nutrients of water from the soil; the rhizoids are used to provide anchorage to their substratum. It is accepted that the chemical composition of the mosses is, in most part, derived from elements that are taken up from atmospheric deposition [8]. The pathway from any source to the moss receptor is stylised in Figure 1.

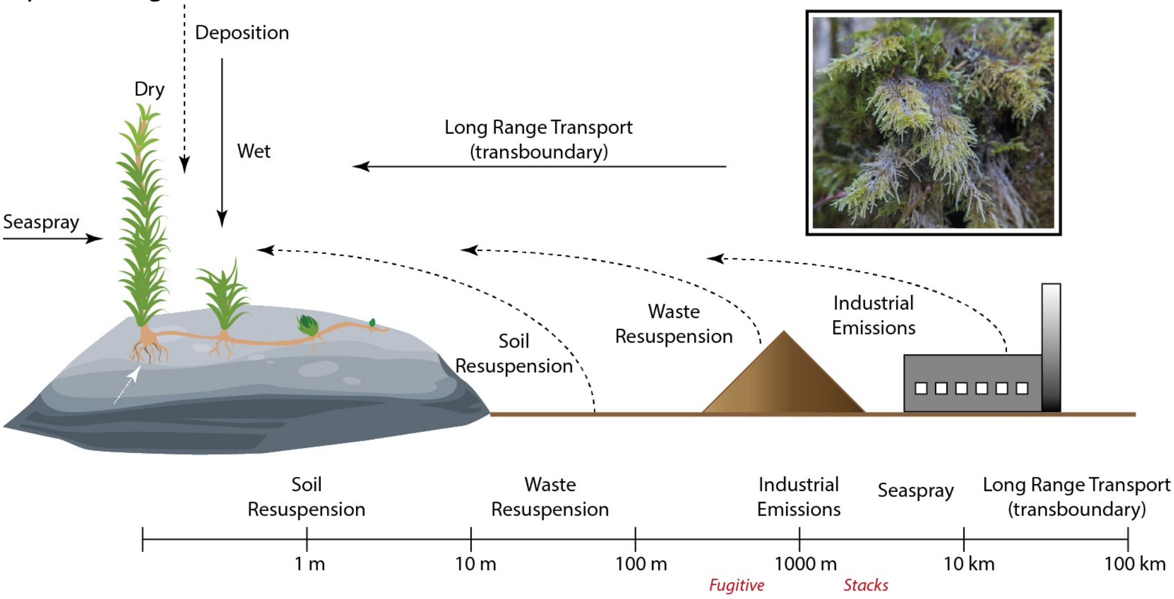

Figure 1. Conceptual model for the mechanism by which elements may be accumulated in mosses together with the range of effect. Industrial emissions may be either fugitive, low-level losses or ducted, thermally enhanced through stacks. It should be noted that there is no uptake through the rhizoids (white arrow) as these are used for anchorage only.

For mosses to reflect the local geological composition, the path length will be short and probably due to soil splash during rainfall events. Waste materials deposited in the environment may be transported through wind-blow and travel greater distances depending on the clastic nature of the deposits. Fugitive emissions from industrial facilities may be in the same class and travel similar distances. Ducted industrial emissions from elevated stacks are likely to have a degree of thermal lift and travel further. Indeed, mosses near a stack may not receive significant input from this source whereas samples located further away may do so. This feature may enable discrimination between fugitive low level emissions and regulated stack emissions (see later section).

3.1 ICP-Vegetation Responses

Responses were received from 10 out of 20 participants (incl. chairman). The response from chairman of ICP-Vegetation is as follows:

As the moss survey is not an approved method for determining absolute air pollutant concentrations or deposition, it is not used in any framework to enforce legislation, but could be used to monitor effectiveness and efficiency of implemented air pollution abatement policies at a high spatial resolution. For EU and UNECE air pollution abatement policies there is a legal *requirement for countries to report official emission data, which are then used in EMEP modelling to estimate air concentrations and deposition.*

On national level, only two countries reported back on use of moss survey around industries:

- Austria reports that authorities have financed moss surveys to monitor seven industrial sites as part of a monitoring programme that included other techniques as well, one industrial site was monitored after an accidental release of pollutants in 1996, and two sites were monitored of the same reason in 2007. In addition, moss monitoring was used in a study of emission from road traffic at 61 sites in Austria (2003-2004).
- Sweden reports that moss surveys are used and performed by different industries to show the impact of their emissions. Boliden Mineral Aktiebolag used moss biomonitoring around Aitik Copper mine in 1995. The results demonstrated that there was an enhancement of 20-25 times the baseline (background used in the report) within an area with a 5 km radius of the works [9].

None of the responding participating countries knew of any use of moss data in any form of legislation.

The reviewed scientific papers tend to fall into three main categories summarised in the next sections. Additionally, there were a small number of manuscripts that investigated biochemical mechanisms for uptake or confounding effects [10] or the effects of deposited elements of metabolic processes [11].

3.2 Source identification and source apportionment in a range of urban and rural locations

A number of authors [12-17] have used moss samples and subsequent statistical analysis to identify the sources of metal contamination and, to a lesser extent, the contribution each source makes to the total load. The typical post-analytical approach has been through the use of principal components analysis (PCA) which might indicate which samples cluster together and which elements behave the same, but it does not quantify how much of each source is in a single sample [18]. The research data have identified that generic industrial emissions can be identified (*e.g.* coal and oil burning [17, 19], metal smelters [13, 20, 21], raw material extraction [22, 23]) along with vehicle emissions [14, 17, 24-26] in the moss samples. In many cases [17, 27-29], it has been concluded that the data could be used for intervention by regulators, but there has been no apparent use so far. This was confirmed by the responses from ICP-Vegetation (3.1). In some cases, specific industrial plants were investigated [17] to better understand the enrichment from coal and oil burning against a regional background that also contained vehicle emissions. While some studies did not target specific industries, they did compare those regions with significant industrial activity with those that could be considered less industrialised [20, 30]. In several cases, the concentrations measured were compared to a “pristine” environment and this was typically samples from northern Norway [31, 32]. It should be noted that none of these studies explicitly set out to “quantify” emissions or to apportion “responsibility” in the data. However, with the use of the correct multivariate statistical tools [18] this would be possible.

3.3 Identification of long-range transport of contaminants

In the Norwegian context [1-7], mosses have been used to show the long range transport of metals from southern Europe reaching the southern part of Norway (*e.g.* Figure 2) and the emissions from Nikel in Russia spreading across northern Norway (Figure 3). In the case of lead (Pb) in Figure 2, the concentrations alone are sufficient to indicate the most impacted regions and the gradient away from the most likely source. It is possible to see, however, other sites in Norway that are relatively enriched compared to the majority of the samples due to local practices.

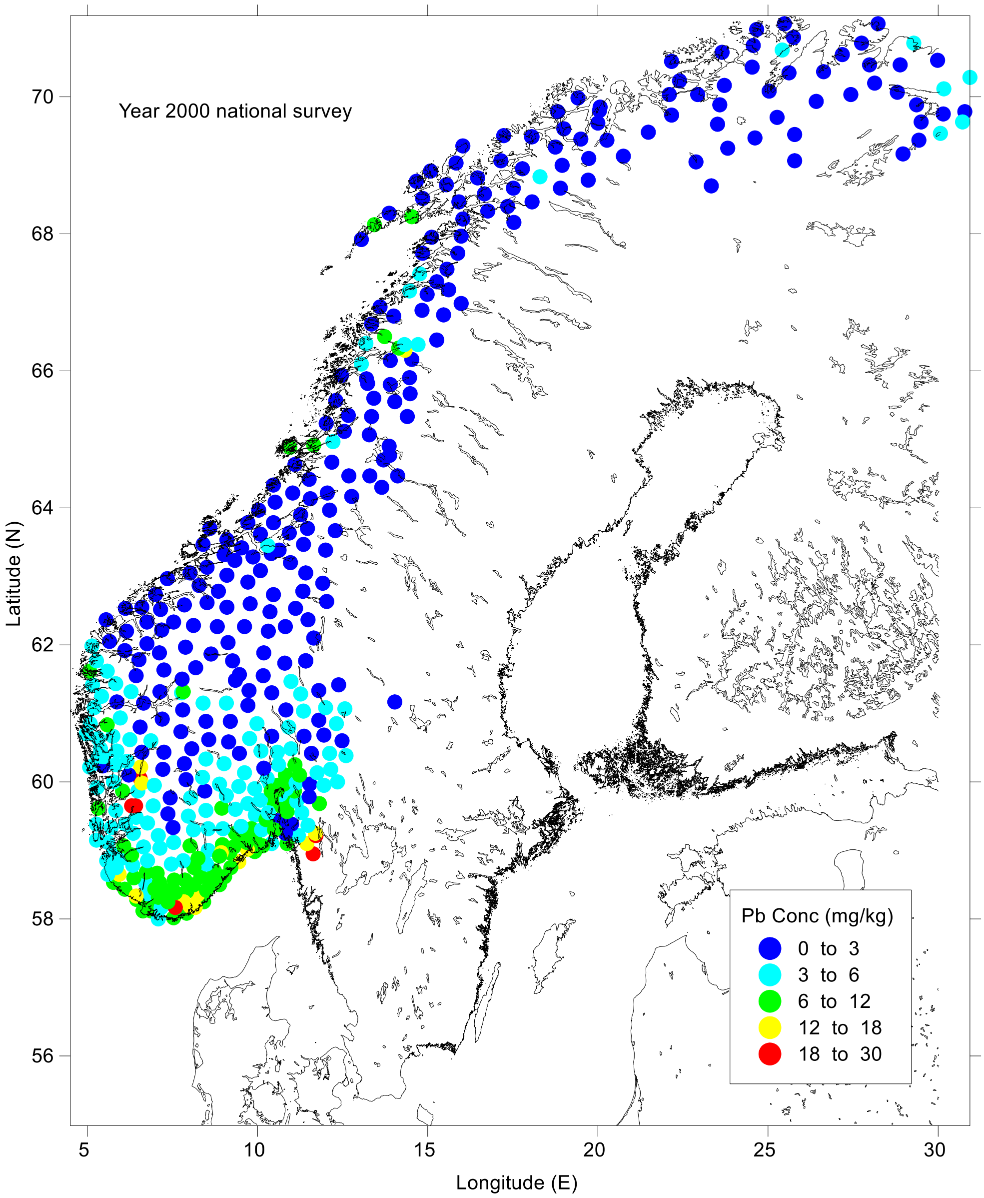

Figure 2. An example of a metal contaminant with multiple sources to the south of Norway reaching southern Norway as a result of long range atmospheric transport. Data from 2000 national survey.

The approach taken with the long range transport from Nikel investigated in Figure 3 develops a more complex chemical signature using several elements. Statistical methods such as partial least squares (PLS) and polytopic vector analysis (PVA) may be used to tease apart the contributions that each of the potential sources makes to the moss [18]. The effect of the Nikel emissions can be distinguished above the baseline condition for 450 km into Norway. It is worth noting that in the results shown in Figure 3, a number of high values shown by the red spots (close similarity to the chemical signature of the emissions from Nikel), can also be seen at industrial sites such as Odda and Mo i Rana. This is consistent across several years and may indicate a generic "metal processing" signature that can be further refined according to the elements being produced.

A few manuscripts discussed the results of analysis of nitrogen and its compounds in moss; in these cases, the moss was being used to determine the nitrogen deposition [33-35] and they concluded that mosses are also useful to estimate the nitrogen deposition to a region.

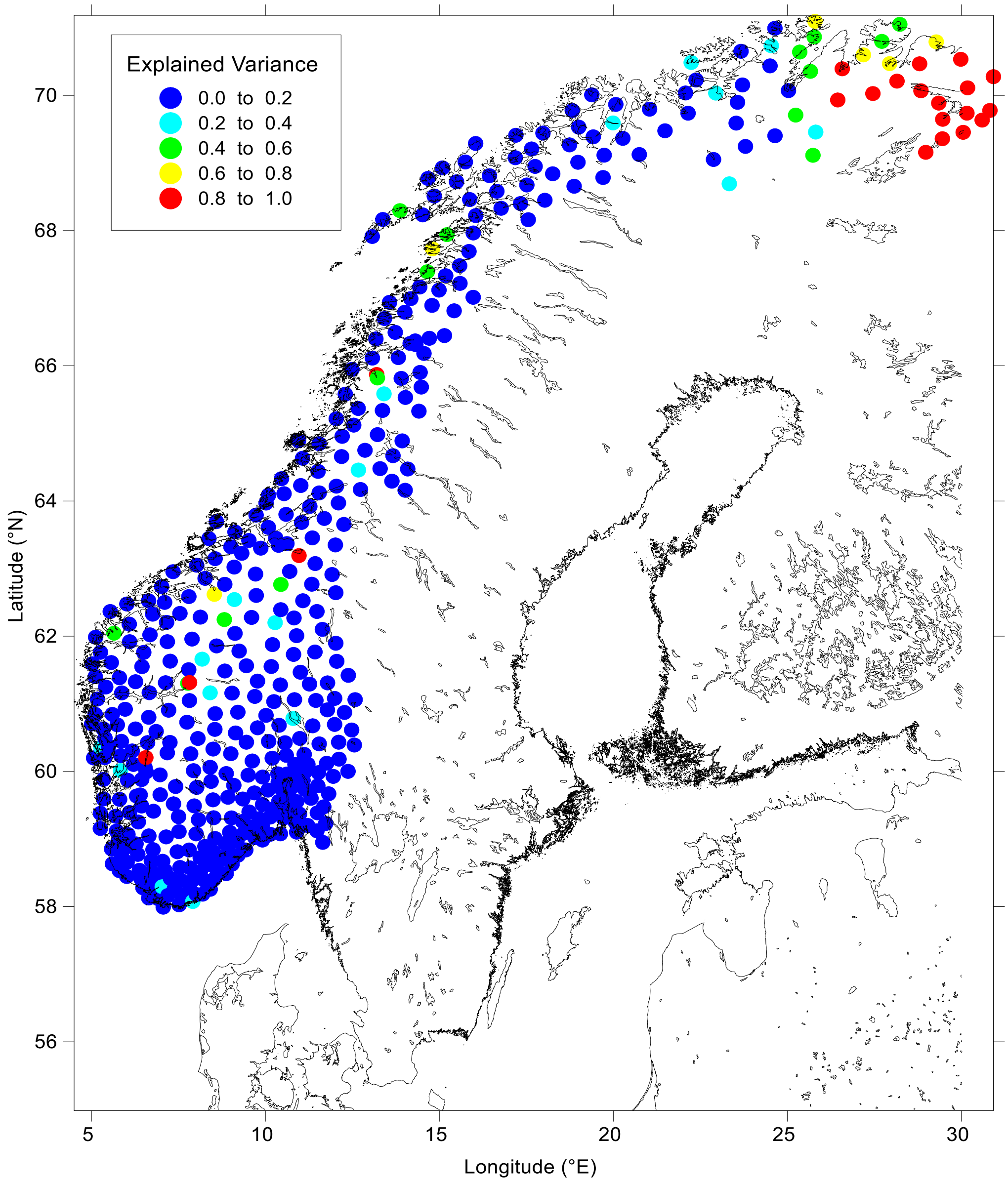

Figure 3. The amount of variance in all the moss samples from the national survey that can be explained by a chemical signature developed from the emissions at Nikel, Russia. (Mudge, unpublished data).

3.4 Industrial emissions near population centres

In a few cases, mosses were used to assess the contamination in urban zones in order to identify "hot spots" although this has been rather sparsely used [36, 37]. Due to the paucity of moss to sample in urban environments, moss bags (see later) were the most common approach to sampling. *In situ* mosses have been used to determine the urban background chemical composition as well [38]. In the urban context, the primary source of metals to the mosses has been vehicle emissions. This compares to geological and long-range sources in more rural environments [24].

4 Approaches to moss use

Mosses may be used as passive samplers in several different ways although it should be noted that comparative studies have indicated that the greatest factor effecting the chemical signature in mosses was the species rather than the source [39]. This has implications when sampling over a large geographic area where the species may change [29]. This is less relevant in Norway where the same species (*Hylocomium splendens*) may be found throughout [40].

Generally, moss methods do not appear to distinguish between contributions from wet and dry deposition, nor will the method provide information on whether each element mainly is associated with small or large particles. However, one manuscript using mosses in forests has suggested that dry deposition may be greater than wet [41] although this could be due to shading. With regard to human health implications, a much more detailed investigation of how each element is distributed between the different size fractions would be necessary.

4.1 Natural mosses

Many studies collect and analyse mosses naturally growing in the region (*e.g.* [28, 42]). This has been the case for the national surveys in Norway.

There are a number of positive reasons for adopting this approach:

- The mosses are acclimated to their growth position with little or no additional stress associated with translocation.
- If the same sample species are collected from the same type of environment, this source of variability is ruled out and only depositional effects are influencing the chemical signature.
- Some mosses such as *Hylocomium splendens* have a recognisable growth interval that integrates deposition over many years. This provides a good measure of the average conditions at this location.
- The sites can easily be resampled at later dates as in the Norwegian surveys.

However, there are also a number of negative factors associated with this approach:

- There may be different moss species in different locations if the geographical region is relatively large. This has been noted as a major factor in determining the chemical signature or concentrations in France [39].
- The moss may not be available at locations where they are needed as part of a geospatial sampling plan. This is especially true in urban and peri-urban environments [17, 36, 43] or locations where mosses do not normally grow [44] and so transplanted mosses may be used instead (see below).

4.2 Transplanted mosses

At times or locations where mosses are unavailable, it is possible to transplant moss from one location to the test locations. However, there are a number of issues related to such practices – the moss needs to be collected from “clean” environments and all samples need to be relatively homogenous before installation [43]. It will take time for the moss to reach equilibrium at the test location and this may depend on the emission rates and periodicity [41].

Transplanted mosses may be used in two different ways.

4.2.1 Moss plus substrate

Naturally growing mosses moved, including their substrate, from less contaminated regions to region under investigation. The moss may then be attached to new locations such as trees and sampled at some time in the future once sufficient deposition has occurred [45]. One of the issues with transplanting on to trees is the potential for shading from deposition and this may have an effect compared to those mosses in open areas.

4.2.2 Moss bags

The more common approach using transplanted moss is through the collection of moss from a pristine location, their placement in bags and hung at the required sampling locations. A number of examples of this approach have been used in urban areas [17, 36, 46] and airports [26].

There are positive reasons for transplanting mosses:

- They can be placed where the sampling design dictates (maybe a function of the location of the industrial sources or human habitation).
- The time of exposure can be controlled and the differences between placement and collection well defined.

However, there are a number of negative factors associated with moss transplanting:

- Mosses have to be collected from the donor location, tested, packed or secured to their substrate, moved to the test locations and then retrieved from those locations after exposure. This needs a significant increase in resources.
- Mosses may be more stressed due to the transplantation process and may have different uptake dynamics compared to acclimatised mosses.
- Mosses in bags may be at risk from petty crime and be lost.
- Devitalised mosses may be used that overcome some of these issues, but these mosses will have different characteristics compared to living mosses and so cross-comparison is not possible.

4.3 Clone mosses

To remove the interspecies variability identified by other researchers [39], one moss has been cloned so that it can be deployed as in 4.2 above [30]. This approach has the advantage of removing the inter- and intra-species variability and providing a low baseline chemical composition in which to identify the enrichment of industrial deposition against. However, the cloning of mosses may be expensive and takes time to produce sufficient material for a sampling campaign.

5 Measuring impacts with mosses

While there have been a significant number of studies mapping the relative effects of emissions and transport using mosses, there have been few studies that link the measured moss signal to that in the atmosphere or in the deposition.

5.1 Impact of emissions on the environment

The concept of using mosses as a bio-*indicator* of impacts is much less developed compared to the widespread use of mosses as bio-*monitors*. There may also be a confusion in the use of the terms [47]. Bio-monitoring is the use of a biological organism such as moss to act as a passive (or active) sampler enabling analysis of the environmental condition. Bio-indicators are organisms that react to a stressor such as atmospheric deposition and the changes to the organism indicate "harm" has occurred. A significant review [35] of the effects of atmospheric deposition on terrestrial biota identified effects of contaminants on mosses. Some of these effects were indirect through changes in the soil chemistry leading to changes in plant species composition that might compete with moss. However, there have been no studies that use mosses to indicate adverse environmental conditions from metals specifically although there have been some studies focusing on nitrogen deposition. These effects might only be measureable at the cellular level only [11, 35].

The environmental consequences of many industrial emissions largely depend on the chemical and physical form of the elements. The uptake of metals by different organisms largely depends on the bioavailability of the metal; in most cases this means enhanced uptake of the more easily soluble forms. The solubility of metal containing particles from a discharge depends strongly on the element itself and the particle size distribution due to surface area effects (smaller particles have a greater surface area per unit mass). A number of other factors will determine solubility including pH and the mineralogical form of the element [48]. Gases and vapour be more "available" through either solubility in precipitation or direct uptake by plants, but they will also be more widely dispersed compared to particulate emissions.

5.2 Impact of emissions on human health

Although several papers suggest action should be taken to improve air quality in respect to human health based on moss analyses, they do not explicitly link either concentrations or chemical signatures to human health conditions. There are a substantial number of confounding factors (diet, smoking, residence time within a particular area, employment, *etc.*) that would make it very difficult to link chemicals measured in mosses to human health effects. A single study published in 2019 has attempted to use health outcomes and establish a link to ambient air metal concentrations using moss data [49]. The authors used a cohort of people who worked for the electricity / gas companies (the GAZEL cohort) as the source of human health outcomes and then linked this to the French national moss monitoring data. Using natural cause deaths along with cardiovascular and respiratory mortality as outcomes, the authors established a significant positive association between the cardiovascular and respiratory mortality and moss-derived Cd and Pb data. The authors indicate that given sufficient highly-resolved data for both moss and human health outcomes, some tentative linkages may be made.

Investigations using the moss methods described in current publications will not provide information on the extent to which a measured element represent a risk to the environment or human health. However, a number of risk ratios may be developed that can indicate the margin of safety against a known measure of toxicity [50]. On the other hand, if the measured concentration of an element in moss is modest, it is possible to suggest it will have little impact. This simplistic approach, however, does not accommodate synergy between elements and compounds.

6 Relating measured concentrations in moss to other factors

The use of mosses as passive bio-monitors is wide spread across Europe and the rest of the world [27, 35, 51] although there are a number of caveats that need to be considered when determining how to interpret and use the data.

6.1 Do mosses reflect the ambient air concentrations?

Considering the mechanisms by which contaminants impact on mosses (Figure 1), there are number of pathways including wet and dry deposition. Contaminants in the atmosphere may be in several different phases including gases and vapours, particles and dissolved in precipitation [52]. The concentrations may vary considerably throughout the day and across the year. Any chemical signature measured in a moss sample will be an integration of these short and long term changes and may represent the annual chemical signature. Thus, moss concentration do not directly reflect ambient air concentrations, nor allow a direct estimation of absolute metal deposition rates in the same manner as is obtained from collecting and analysing precipitation. However, an attempt has been made to relate moss concentration with wet deposition data. A study by Berg et al. [53] compared wet deposition data to moss concentrations in pristine areas and used regression equations to transform moss concentration to absolute deposition rates. The authors established a correlation between wet deposition and moss concentration, and significant positive correlations were obtained for elements subjected to long-range transport such as V, As, Se, Mo, Sb, Tl and Pb, whereas elements typically associated with sea-spray and soil dust show a poor correlation. Based on regression equations, calibration factors for the transformation from moss concentrations to absolute deposition have been developed for the long-range transported elements. It must be noted that this method is only applicable to remote inland locations with minor influence of dry deposition. This excludes industrial sites, populated areas and coastal areas.

It should be noted that meteorological conditions will have significant effects on the elements that may be taken up by the moss: If a potential source is upwind of the moss for most of the time, it will make a bigger contribution than a potentially nearer source that is downwind most of the time. In all analytical campaigns, care should be taken to understand the effect of inter-sample distance related to source and the predominant dispersion direction mediated by the wind and topography (see Figure 1 for range of effects).

Sampling programmes that use natural mosses (see 4.1) tend to take portions of the moss that represent growth from the previous 1-2 years [1-7]. If mosses are used for assessment of industrial emissions [20], the location of samples relative to the potential sources is of key importance. Fugitive or diffuse emissions that are generally not measured or reported may contribute to moss close to the works, but less so at the more remote samples. Conversely, ducted emissions with an elevated thermal lift may not impact moss near the works, but will have a greater influence at the more remote locations. This is shown schematically in Figure 4. It is possible, however, that this differentiation may be a mechanism to semi-quantify the diffusive emissions if the ducted emissions are known. By using a multivariate statistical method such as polytopic vector analysis (PVA), the contribution from each source may be calculated and mapped. By integrating the footprint of each source and comparing the measured ducted emissions to the other signatures, the relative magnitude of the diffuse emissions may be calculated.

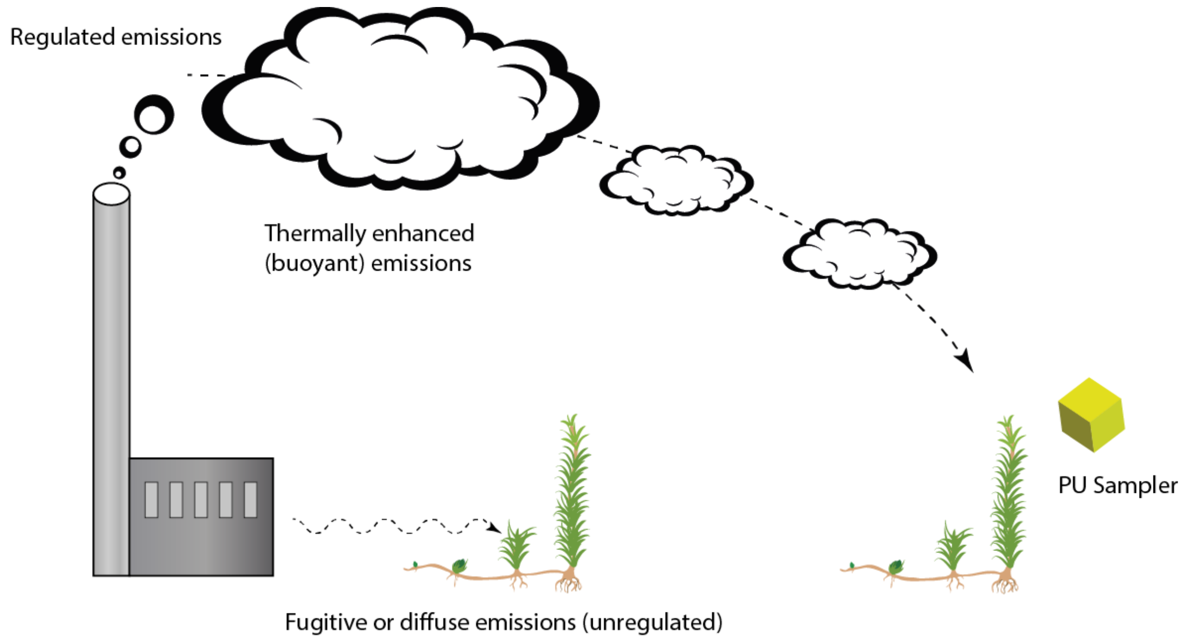

Figure 4. Potential mechanisms by which moss near industrial facilities may acquire discharged elements. While the regulated route may be quantified to some degree, the fugitive or diffuse routes may not be. Polyurethane (PU) foam samplers may be used to collect ambient air samples for short-term exposures.

One of the commonly reported sources of metals in mosses measured in urban areas and near roads is vehicle emissions [17, 22, 24-26, 46, 54]. Different elements are used to determine the contributions (*e.g.* Pb, Cu, Fe and Sb) depending on the choice of the investigators. The larger the dataset collected, however, enables greater source differentiation and a greater range of sources that may be characterised in complex, multi-industry, environments. There are conditions when too many elements blur the differentiation, but it is always possible to remove them and it is much harder to go back and add them in later.

Vehicle emissions tends to be reported as the largest contributor to mosses in urban and peri-urban areas in the absence of specific industrial facilities. When there are known sources of emissions such as coal fired power stations, these often dominate in the immediate vicinity.

6.2 Do mosses indicate the emissions inventory from industries?

Mosses integrate the emissions with time and, if the source signal is variable, this will be blurred in the moss. Mosses provide a chemical signature that can be used to identify a range of emissions and their relative strengths, if correctly sampled (see above). Along with the chemical signature developed from the proportions of each metal in the samples, it is possible to use stable isotopes of selected elements to differentiate between sources. One such example has been the use of stable isotopes of lead (^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb) as these may be linked to specific industrial sources and the raw materials they use [55]. However, these approaches tend to identify the source and its relative importance in any one particular sample, but not the total emissions inventory from a specific source.

A review by Bargagli [27] specifically investigated the biomonitoring of mercury using moss and lichen. It was clear that there are gradients in concentration and signature away from known point sources although the range of effects was within the first few kilometres only; this is consistent with the conceptual model in Figure 1. In his review, Bargagli indicated that the concentrations in the moss were a function of many parameters including the bioavailability of the element, the deposition rate, the pH of the precipitation, metabolic activity of the moss, drought or snow cover. Additionally for some elements there may be a bidirectional flux with elements lost from the moss after initial uptake.

This review paper prompted a quick review of the mercury in Norwegian moss samples for 2010 and 2015. The distributions can be seen in Figure 5 although there is a significantly reduced coverage in 2015. Investigation of the hot spots using maps indicates a potential linkage to both known industrial activity and at unexpected sites such as open cast mineral and sand extraction locations. Given the short path length between source and receptor identified by Bargagli [27], this may warrant more investigation.

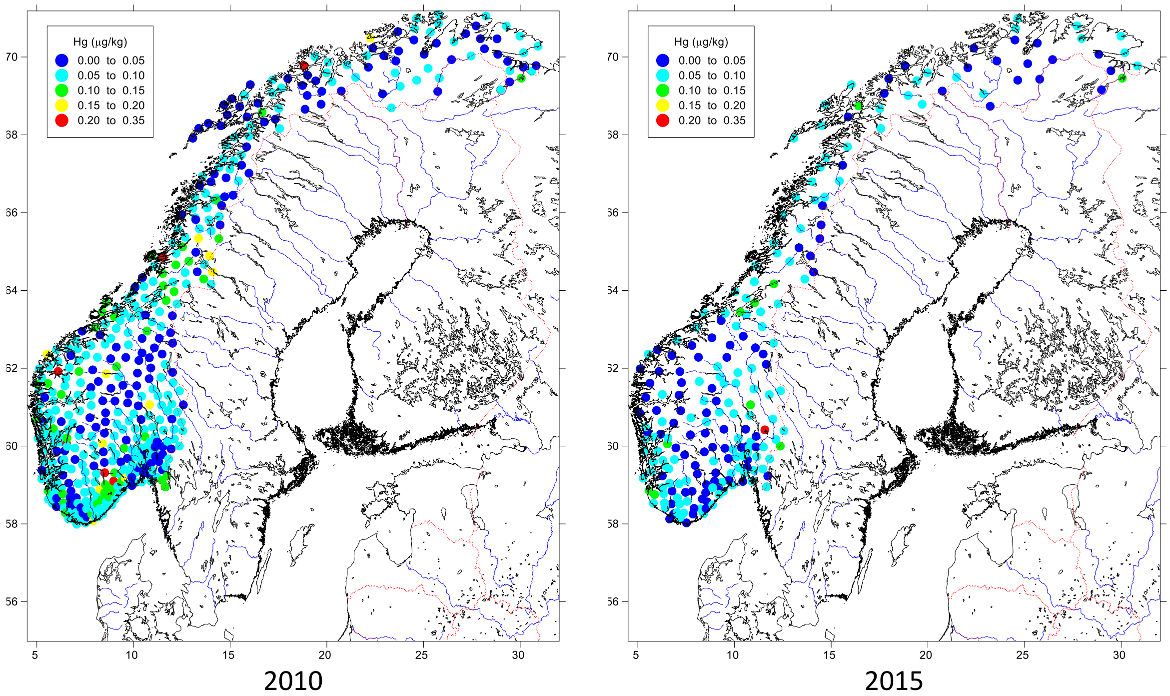

Figure 5. The concentration of Hg in moss from the Norwegian national surveys of 2010 and 2015.

6.3 Do mosses indicate the potential uptake in other biota?

No explicit studies have been identified that test the linkage between elements in mosses and in other biota. There are data from independent studies in Northern Norway that have measured similar suites of elements in wild foods, but paired samples with mosses have not been taken.

Mosses will provide a measure of the elements available for uptake and the relative strength of that signal, but may not reflect the chemical signature in biota, especially those animals that roam across the region.

7 Recommendations

In light of this review, it is clear that there are a number of unanswered questions regarding how mosses may be used in a more regulatory context. The majority of investigations reported to date have simply indicated which sources have been impacting a site; this is only a small fraction of what could be achieved if a series of well-designed studies are conducted with the right post-analytical tools. There are a number of enabling projects that could be conducted using existing data to an extent and this would inform future investigations.

7.1 Interpretation of historical results around Norwegian industrial sites

NILU would be able to conduct a thorough analysis of the existing data collected around the industrial locations of Norway and the country-wide survey which has highlighted hot-spots that may need further investigation (*e.g.* Hg from sand extraction sites, Mo from the closed mines). This uses existing data and maximises the return on the investment in its collection and analysis. At present, the second stage on source identification and apportionment shown in Figure 6 has not been completed. Once done, further knowledge shown in blue in Figure 6 would be obtained. The outputs would include maps of specific emissions from industries as well as the more widely dispersed sources. This would also provide a basis for further work to link quantified emissions with moss-derived concentrations.

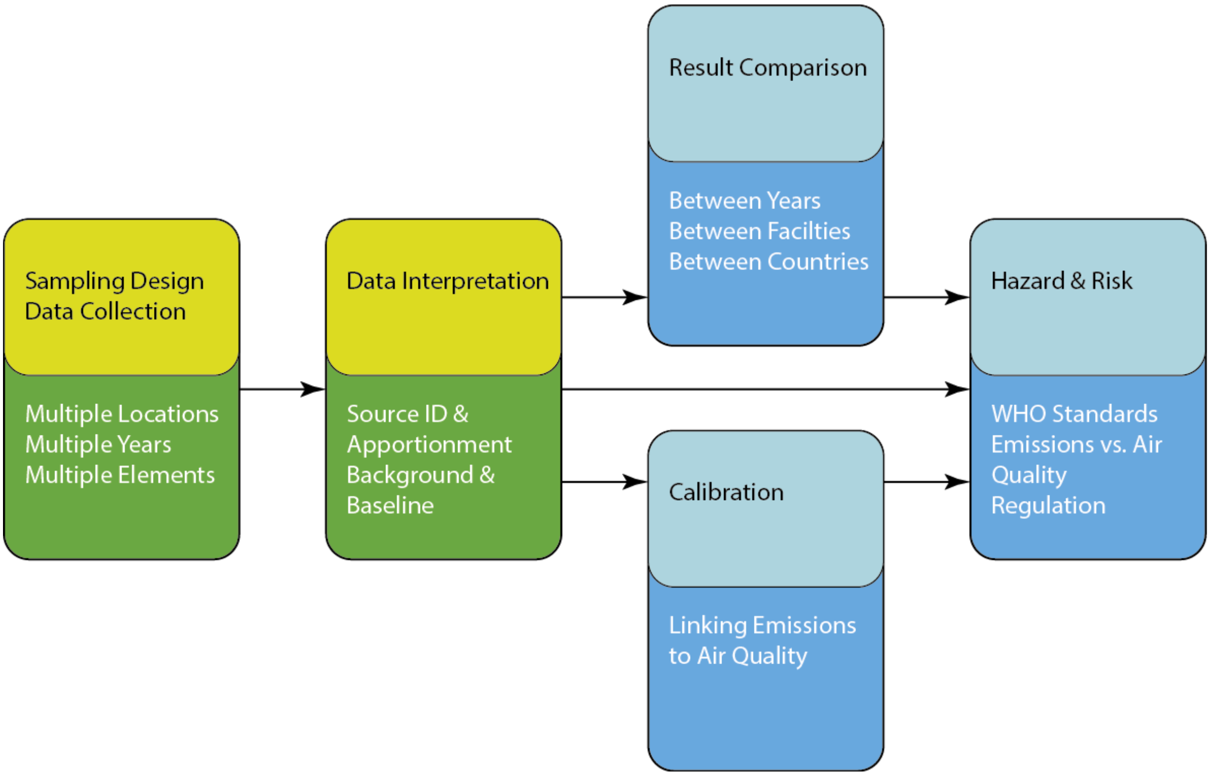

Figure 6. Data and knowledge flow to interpret the existing elemental composition of mosses around industrial facilities. At this present moment, stage 2, the data interpretation, has not been conducted.

7.2 Comparative evaluation of metals in selected biota from Northern Norway

Mosses are very good passive collectors for airborne metals although how these data might relate to metals in other food items is not known. This could be achieved by comparing existing data on metals in biota with metals in moss to establish the degree of linkage. This prediction could be for concentrations and/or signatures. The outcome from this study would be an answer to the question - can mosses be used as an indicator of likely body burden in wild food items? The degree of linkage between the different items can be investigated and tested on a separate suite of samples. Some data from Northern Norway already exist that could be used in the formulation of a sampling / analysis plan. It was also indicated that mosses are particularly good at biomonitoring in alpine or polar areas where they may be the dominant ground cover [27].

7.3 Establishing the linkage between air quality and moss chemical signatures

One of the key goals for a regulator might be a cost-effective mechanism to determine the ambient air quality over an extended period. A series of experiments could be conducted to determine the linkage between ambient air quality and moss chemical composition. This would involve installing high frequency (days to weeks) active and passive samplers near known sources that can be identified with the current moss data (Figure 4). The rate of uptake into moss can be measured along with the time to equilibrium. Factors may be developed that enable an estimate of the emissions to be predicted. This would need to be done at several different sites with different emission types. Also, we would determine the role of deposition type (dry and wet) and altitude on uptake.

7.4 Using mosses as a proxy for other (wild) food items

When collecting samples for the next national survey, samples of selected wild foods / berries could additionally be collected at the same locations to investigate the linkage between species. Models to develop a link between the metadata for the sites will help understand the mechanisms by which the metals may be accumulated. This would provide an insight into the potential for human exposure through wild food consumption.

7.5 Systematic survey of industrial emissions (ducted and fugitive) with mosses

It is possible to conduct a systematic survey of the chemical signatures in mosses around known facilities that have emissions, either ducted through stacks or from wind blow / fugitive routes (Figure 7). The mosses must be sampled according to a correctly designed programme so that the outcomes can be achieved – this will be different for each facility to be investigated. This may mean moss bags instead of natural mosses in some locations although the preferred mechanism would be natural *in situ* mosses. If samples are collected close to potential fugitive emission sites as well as in the wider environment, the contributions from each source can be calculated using PVA. If the emissions through the stacks are known, this can be used to calculate the quantity lost from sites through diffuse / fugitive emissions.

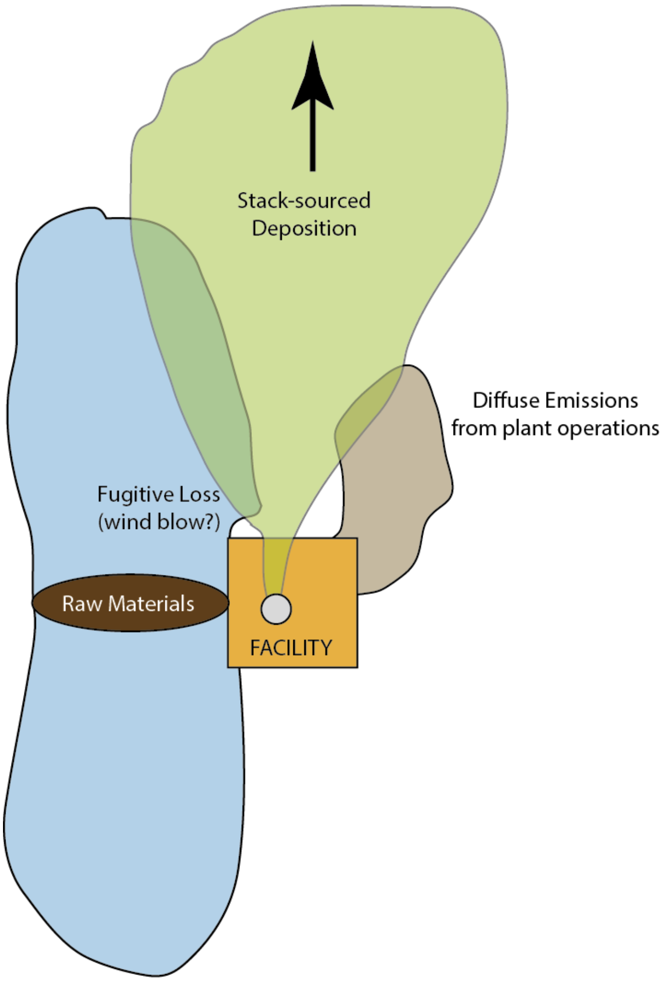

Figure 7. A conceptual model of how emissions from several sources from a single industrial facility may mingle offsite.

8 Conclusions

There is no doubt that mosses are good passive samplers of airborne contaminants and can provide information on the chemical signature of local depositions. This is recognised by the ICP-Vegetation group and through the numerous studies detailed above. The aspect that has been lacking until recently, has been the ability to relate the chemical data measured in mosses to the ambient air quality or emissions from selected industries. Reassessment of the national data from Norway has been able to clearly identify "locations of interest" due to the enrichment of either individual elements or more complex chemical signatures comprised of several elements. However, the sampling design for the national survey is not suitable to investigate the emissions from individual industries where a different inter-sample distance is needed. The data from the industrial surveys since 2000 are available although they have not been re-assessed to produce emissions maps. This could be completed on the existing data as suggested in 7.1.

A single 2019 paper [49] has made an attempt to link the elemental composition of mosses to human health outcomes. This has indicated that making such a link is possible and, as a leader in the use of mosses in Europe, something that could be conducted across Norway. With regard to assessing the effects of ducted and fugitive emissions from industries on the local environment, a sampling and analytical programme needs to be designed that specifically addresses this question. There is a sequence of stages needed to ensure a good outcome from reviewing the known emissions, modelling the most likely deposition sites, designing the moss sampling programme in response to the model results, sampling and analysis and, critically, the post-analytical treatment of the data to distinguish between sources and calculate the relative source strength. Determining cause and effect requires careful consideration and a substantial dataset to remove or reduce the confounding factors. Such a project is possible with the right investment.

9 References

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55. E. Schnyder, M. Strok, Z. Kosonen, M. Skudnik, D. Mazej, Z. Jeran and L. Thoni, Lead concentrations and stable lead isotope ratios in moss in Slovenia and Switzerland, *Ecological Indicators*, 2018, **95**, 250-259.

10 Appendix – Summary of reviewed papers

Number	Reference	Geographical region / Approach	Summary
17	I. Zinicovscaia, M. Anicic Urosevic, K. Vergel, E. Vieru, M. V. Frontasyeva, I. Povar and G. Duca, ACTIVE MOSS BIOMONITORING OF TRACE ELEMENTS AIR POLLUTION IN CHISINAU, REPUBLIC OF MOLDOVA, *Ecological Chemistry and Engineering S-Chemia I Inzynieria Ekologiczna S*, 2018, **25**, 361-372.	Moss bags in an urban environment of Moldova	Trace element deposition around Chisinau as a biomonitoring exercise. Identified significant enrichment around the power plant (coal and oil) and from vehicle emissions.
41	G. Kosior, A. Dolhanczuk-Srodka and Z. Ziembik, THE USE OF MOSS Pleurozium schreberi (Brid.) Mitt. AS BIOINDICATOR OF RADIONUCLIDE CONTAMINATION IN INDUSTRIAL AREAS OF UPPER SILESIA, *Ecological Chemistry and Engineering S-Chemia I Inzynieria Ekologiczna S*, 2017, **24**, 19-29	Transplanted mosses in Upper Silesia, Poland	Identified a number of sources based on the radionuclide decay series and known emissions profiles. Fossil fuel burning was the major source of ^{210}Pb.
35	L. P. Wright, L. M. Zhang, I. Cheng, J. Aherne and G. R. Wentworth, Impacts and Effects Indicators of Atmospheric Deposition of Major Pollutants to Various Ecosystems - A Review, *Aerosol and Air Quality Research*, 2018, **18**, 1953-1992	Review of the atmospheric deposition of selected pollutants	With regard to moss, noted that nitrogen deposition may have an effect on mosses and that they also accumulated PAHs.
19	J. A. Galhardi, R. Garcia-Tenorio, I. D. Frances, D. M. Bonotto and M. P. Marcelli, Natural radionuclides in lichens, mosses and ferns in a thermal power plant and in an adjacent coal mine area in southern Brazil, *Journal of Environmental Radioactivity*, 2017, **167**, 43-53.	Targeted investigation of coal-fired power plant in Brazil using *in situ* mosses, lichen and ferns	Mosses were the better accumulator of the lead but only four samples analysed. The mining of coal was the major source of the measured radionuclides.
30	A. Di Palma, D. C. Pardo, V. Spagnuolo, P. Adamo, R. Bargagli, D. Cafasso, F. Capozzi, J. R. Aboal, A. G. Gonzalez, O. Pokrovsky, A. K. Beike, R. Reski, M. Tretiach, Z. Varela and S. Giordano, Molecular and chemical characterization of a Sphagnum palustre clone: Key steps towards a standardized and sustainable moss bag technique, *Ecological Indicators*, 2016, **71**, 388-397.	Development of a moss clone	A moss clone was developed that has a low intrinsic metal content to enable low-levels of contaminants to be detected by ICP-MS.

Number	Reference	Geographical region / Approach	Summary
29	E. Lequy, S. Sauvage, X. Laffray, S. Gombert-Courvoisier, A. Pascaud, L. Galsomies and S. Leblond, Assessment of the uncertainty of trace metal and nitrogen concentrations in mosses due to sampling, sample preparation and chemical analysis based on the French contribution to ICP-Vegetation, *Ecological Indicators*, 2016, **71**, 20-31.	French national survey (1996, 2000, 2006 and 2011) with 449 – 536 sites.	The uncertainty associated with moss analyses was considered and how a sampling programme should be designed to take these errors into account.
55	E. Schnyder, M. Strok, Z. Kosonen, M. Skudnik, D. Mazej, Z. Jeran and L. Thoni, Lead concentrations and stable lead isotope ratios in moss in Slovenia and Switzerland, *Ecological Indicators*, 2018, **95**, 250-259	Pb in mosses from Swiss and Slovenian samples. Used MC-ICP-MS.	The stable isotopes of Pb were used to show the different sources although this was not an industrially targeted programme.
22	H. J. Bing, Y. H. Wu, J. Zhou and H. Y. Sun, Biomonitoring trace metal contamination by seven sympatric alpine species in Eastern Tibetan Plateau, *Chemosphere*, 2016, **165**, 388-398	Limited metals by ICP-MS in trees and moss from Tibet	Used a combination of metals and isotopes to associated the distribution to mining operations, coal usage and vehicle emissions.
10	M. Renaudin, S. Leblond, C. Meyer, C. Rose and E. Lequy, The coastal environment affects lead and sodium uptake by the moss Hypnum cupressiforme used as an air pollution biomonitor, *Chemosphere*, 2018, **193**, 506-513.	Coastal French sites investigating the link between Na and Pb – are there coastal effects?	Due to competition for binding sites, close to the coast where Na is high, Pb may be underrepresented.
27	R. Bargagli, Moss and lichen biomonitoring of atmospheric mercury: A review, *Science of the Total Environment*, 2016, **572**, 216-231	Review of Hg in mosses	Clearly showed the change in Hg with distance from known sites with sharp decrease within the first km. Confirmation of the CSM presented in Figure 1. Geothermal power plants source. Uptake by moss may be influenced by bioavailablity of Hg, deposition rate pH and metabolic activity. [Also snow cover and drought may be important in Norway]

Number	Reference	Geographical region / Approach	Summary
39	E. Lequy, N. P. A. Saby, I. Ilyin, A. Bourin, S. Sauvage and S. Leblond, Spatial analysis of trace elements in a moss bio-monitoring data over France by accounting for source, protocol and environmental parameters, *Science of the Total Environment*, 2017, **590**, 602-610	Used the French national datasets. Focussed on six metals.	Major factor influencing variation with data was species. [Not relevant to Norway as the same species used throughout]
28	A. Klos, Z. Ziembik, M. Rajfur, A. Dolhanczuk-Srodka, Z. Bochenek, J. W. Bjerke, H. Tommervik, B. Zagajewski, D. Ziolkowski, D. Jerz, M. Zielinska, P. Krems, P. Godyn, M. Marciniak and P. Swislowski, Using moss and lichens in biomonitoring of heavy-metal contamination of forest areas in southern and north-eastern Poland, *Science of the Total Environment*, 2018, **627**, 438-449	500 moss and lichen samples for seven metals in Polish forests.	Seasonal changes with some metals and gradients away from known sources of metals. Confirms ability to indicate industrial emissions.
11	D. L. Scott, R. L. Bradley, J. P. Bellenger, D. Houle, M. J. Gundale, K. Rousk and T. H. DeLuca, Anthropogenic deposition of heavy metals and phosphorus may reduce biological N-2 fixation in boreal forest mosses, *Science of the Total Environment*, 2018, **630**, 203-210	Not environmental tracking of sources	Demonstrates that P and Mo may directly affect the metabolism of moss.
43	T. Milicevic, M. A. Urosevic, G. Vukovic, S. Skrivanj, D. Relic, M. V. Frontasyeva and A. Popovic, Assessment of species-specific and temporal variations of major, trace and rare earth elements in vineyard ambient using moss bags, *Ecotoxicology and Environmental Safety*, 2017, **144**, 208-215	Moss bags in vineyards. 41 elements in Serbia	Demonstrated that vineyards were a source of selected metals, potentially due to treatments. Increased with time and vineyards a source of diffuse emissions.
34	Z. Kosonen, A. Thimonier, E. Schnyder and L. Thoni, Nitrogen concentration in moss compared with N load in precipitation and with total N deposition in Switzerland, *Environmental Pollution*, 2018, **239**, 169-178	Used moss to show increased deposition of nitrogen. 24 sites in Switzerland	Showed mosses good for long term monitoring and that there would be a benefit in including N in future Norwegian studies.
15	J. L. Liu, X. Y. Bi, F. L. Li, P. C. Wang and J. Wu, Source discrimination of atmospheric metal deposition by multi-metal isotopes in the Three Gorges Reservoir region, China, *Environmental Pollution*, 2018, **240**, 582-589	36 moss samples from eight sites in China. 18 metals.	Used stable isotopes of Pb to differentiate between sources.

Number	Reference	Geographical region / Approach	Summary
44	J. Arndt, S. Calabrese, W. D'Alessandro and B. Planer-Friedrich, Using mosses as biomonitors to study trace element emissions and their distribution in six different volcanic areas, *Journal of Volcanology and Geothermal Research*, 2017, **343**, 220-232	Global study of moss bags sited near volcanoes.	Demonstrated usefulness of moss bags to monitor volcanic emissions.
36	J. Limo, P. Paturi and J. Makinen, Magnetic biomonitoring with moss bags to assess stop-and-go traffic induced particulate matter and heavy metal concentrations, *Atmospheric Environment*, 2018, **195**, 187-195	Moss bags at traffic lights in Finland	Used magnetic properties to show metal accumulation due to traffic.
13	P. Cowden and J. Aherne, Assessment of atmospheric metal deposition by moss biomonitoring in a region under the influence of a long standing active aluminium smelter, *Atmospheric Environment*, 2019, **201**, 84-91.	Canadian study, 61 samples for 13 metals.	Showed clear dispersion away from aluminium smelters. Concentrations dependant on distance.
38	M. A. Urosevic, G. Vukovic, P. Jovanovic, M. Vujicic, A. Sabovljevic, M. Sabovljevic and M. Tomasevic, Urban background of air pollution: Evaluation through moss bag biomonitoring of trace elements in Botanical garden, *Urban Forestry & Urban Greening*, 2017, **25**, 1-10	Mass bags in Serbia	Study on urban background [baseline] in Serbian city
25	E. Rota, B. Braccino, R. Dei, S. Ancora and R. Bargagli, Organisms in wall ecosystems as biomonitors of metal deposition and bioavailability in urban environments, *Environmental Science and Pollution Research*, 2018, **25**, 10946-10955	Urban metals using mosses on walls as biomonitors.	Was able to show mosses collected metals to help define chemical signature of the city baseline.
42	S. Allajbeu, F. Qarri, E. Marku, L. Bekteshi, V. Ibro, M. V. Frontasyeva, T. Stafilov and P. Lazo, Contamination scale of atmospheric deposition for assessing air quality in Albania evaluated from most toxic heavy metal and moss biomonitoring, *Air Quality Atmosphere and Health*, 2017, **10**, 587-599	Nine metals in 44 samples in Albania	Concluded that the siting of samples is important when trying to differentiate between long-range and local sources.

Number	Reference	Geographical region / Approach	Summary
31	S. Berisha, M. Skudnik, U. Vilhar, M. Sabovljevic, S. Zavadlav and Z. Jeran, Trace elements and nitrogen content in naturally growing moss Hypnum cupressiforme in urban and peri-urban forests of the Municipality of Ljubljana (Slovenia), *Environmental Science and Pollution Research*, 2017, **24**, 4517-4527	44 sites in Slovenia. Reference Norwegian national survey data as a "background" value.	Urban sources identified as traffic, heating and soil.
32	S. Shetekauri, O. Chaligava, T. Shetekauri, A. Kvlividze, T. Kalabegishvili, E. Kirkesali, M. V. Frontasyeva, O. E. Chepurchenko and V. A. Tselmovich, Biomonitoring Air Pollution Using Moss in Georgia, *Polish Journal of Environmental Studies*, 2018, **27**, 2259-2266	36 moss samples analysed for 47 elements in Georgia. Also used Norwegian data for background.	Showed industrial sources but did not establish a source – pathway – receptor gradient.
23	J. A. G. Corisco, J. Mihalik, M. J. Madruga, M. I. Prudencio, R. Marques, M. Santos and M. Reis, Natural Radionuclides, Rare Earths and Heavy Metals Transferred to the Wild Vegetation Covering a Phosphogypsum Stockpile at Barreiro, Portugal, *Water Air and Soil Pollution*, 2017, **228**.	Waste ponds in Portugal.	Showed mosses a good collector of deposition and wind-blown matter from waste ponds.
46	R. Hu, Y. Yan, X. L. Zhou, Y. N. Wang and Y. M. Fang, Monitoring Heavy Metal Contents with Sphagnum Junghuhnianum Moss Bags in Relation to Traffic Volume in Wuxi, China, *International Journal of Environmental Research and Public Health*, 2018, **15**	Moss bags in China. Five metals at five sites two seasons	Major source was vehicle emissions
54	Y. B. Jiang, M. Fan, R. G. Hu, J. S. Zhao and Y. P. Wu, Mosses Are Better than Leaves of Vascular Plants in Monitoring Atmospheric Heavy Metal Pollution in Urban Areas, *International Journal of Environmental Research and Public Health*, 2018, **15**	12 metals by ICP in China	Showed higher concentrations in the moss near known sources (roads and industry)
41	G. Kosior, A. Samecka-Cymerman and A. Brudzinska-Kosior, Transplanted Moss Hylocomium splendens as a Bioaccumulator of Trace Elements from Different Categories of Sampling Sites in the Upper Silesia Area (SW Poland): Bulk	Transplanted mosses at 15 sites in Poland.	Compared different regions with industrial and residential sources. Showed dry deposition was more important than wet depo.

Number	Reference	Geographical region / Approach	Summary
	and Dry Deposition Impact, *Bulletin of Environmental Contamination and Toxicology*, 2018, **101**, 479-485		
24	R. Liu, Z. H. Zhang, J. C. Shen and Z. H. Wang, Analysis of metal content and vertical stratification of epiphytic mosses along a Karst Mountain highway, *Environmental Science and Pollution Research*, 2018, **25**, 29605-29613	Epiphytic mosses along mountain highway in China	Differentiated sources into 1. Geological, vehicles, tyre wear, Pb emissions with historic sources
45	N. S. Rogova, N. K. Ryzhakova and A. L. Borisenko, Effect of placement conditions for active monitoring of trace element with the epiphytic moss, *Environmental Monitoring and Assessment*, 2018, **190**	Transplanted mosses to trees in Russia	A comparison of the methods for sample collection rather than source tracking
25	E. Rota, B. Braccino, R. Dei, S. Ancora and R. Bargagli, Organisms in wall ecosystems as biomonitors of metal deposition and bioavailability in urban environments, *Environmental Science and Pollution Research*, 2018, **25**, 10946-10955	Mosses on urban and suburban walls in Italy	Answering the hypothesis that wall grown mosses can act as passive samplers for Pb, Zn and Cd derived from traffic.
33	E. A. Diaz-Alvarez and E. de la Barrera, Characterization of nitrogen deposition in a megalopolis by means of atmospheric biomonitors, *Scientific Reports*, 2018, **8**.	Mosses in Mexico to track nitrogen deposition	Used a combination of [N] and stable isotopes to track deposition.
26	G. Vukovic, M. A. Urosevic, S. Skrivanj, K. Vergel, M. Tomasevic and A. Popovic, The first survey of airborne trace elements at airport using moss bag technique, *Environmental Science and Pollution Research*, 2017, **24**, 15107-15115	Used moss bags placed around an airport to track emssions in Serbia	Greatest enrichment was found in the car park rather than the runway.

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ISBN: 978-82-425-2991-6
ISSN: 2464-3327

Norsk institutt for luftforskning
Norwegian Institute for Air Research