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Indoor monitoring of heavy metals and NO_2 using active monitoring by moss and Palmes diffusion tubes

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Abstract

Background: Indoor pollution is a real threat to human health all over the world. Indoor pollution derives from indoor sources (e.g. smoking, gas stoves, coated furniture) as well as from outdoor sources (e.g. industries, vehicles). Long-term monitoring measurements in indoor environments are missing to a large extent due to a lack of simple to operate measuring devices. Mosses proved well as biomonitors in hundreds of studies. Nevertheless, indoor use has been extremely scarce. Therefore, this study aimed to determine indoor and outdoor pollution by active biomonitoring using moss as well as NO_2 samplers to analyse outdoor and indoor levels of pollution. We exposed moss (*Pleurozium schreberi*) for 8 weeks indoors and outdoors in 20 households in the city of Girona, Spain. Al, Cr, Cu, Zn, Sn, Cd, Pb, Mo, and Sb were analysed by moss-samplers. Additionally, NO_2 was measured with Palmes diffusion tubes.

Results: Compared to the pre-exposure analysis, concentrations of almost all elements both on indoor and outdoor mosses increased. Except for Cd, all metals and NO_2 had, on average, higher concentrations in outdoor mosses than at corresponding indoor sites. However, some 20% of the samples showed inverse patterns, thus, indicating both indoor and outdoor sources. Indoor/outdoor correlations of elements were not significant, but highest for markers of traffic-related pollution, such as Sn, Sb, and NO_2. The wide range of indoor–outdoor ratios of NO_2 exemplified the relevance of indoor sources such as smoking or gas cooking. Though mostly excluded in this study, a few sites had these sources present.

Conclusions: The study at hand showed that moss exposed at indoor sites could be a promising tool for long-time biomonitoring. However, it had also identified some drawbacks that should be considered in future indoor studies. Increments of pollutants were sometimes really low compared to the initial concentration and therefore not detectable. This fact hampers the investigation of elements with low basic element levels as, e.g. Pt. Therefore, moss with real low basic levels is needed for active monitoring, especially for future studies in indoor monitoring. Cloned material could be a proper material for indoor monitoring yet never was tested for this purpose.

Keywords: Active monitoring, Heavy metals, Moss-samplers, NO_2-samplers, Outdoor/indoor levels of pollution, Outdoor/indoor fluxes

Background

Discussions on the impact of indoor pollution on human health have raised many questions. Leading organizations in the field of health-care have listed indoor pollution as a major cause of concern [1–4]. Long-term monitoring measurements in indoor environments are largely missing due to a lack of adequate measuring devices. People, especially Europeans, spend more

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than half of the time at home, indoors, with another 8–9 h in other indoor environments. Thus, exposure to indoor pollution from both indoor and outdoor sources is a huge health concern [5, 6]. In general, the range of pollutants originating from indoor sources is broad, including environmental tobacco smoke (ETS), cooking with gas, candles, or emissions from furniture and other material, to name a few. Indoor sources for heavy metals, in particular, include, e.g. batteries, water pipes, and combustion processes of various kinds [6, 7]. Significant outdoor sources are traffic and industries [8]. There are several studies on the transport from outdoor sources into indoor environments [6], however many of these models fail as quantitative measurements in indoor environments are inadequate. This failure is mainly due to short periods of observation, which are provided by technical measures [9].

Technical measurements based on chemical–physical methods using stationary or mobile automatic gauges have been the most important source of information about atmospheric pollution, especially in indoor environments [10]. Most of the technical measurements, especially in indoor environments last only for a few hours or days. Biomonitors have provided reliable data on outdoor air pollution for decades [11]. Besides yielding information on the toxicity of chemical substances and the health status of the environment, they can provide valuable information on quantities of deposited hazardous agents. In many investigations, biomonitors were favoured over technical measurements because of their easy handling and economic advantages. Furthermore, biomonitoring is a valid and cost-efficient tool for long-term monitoring.

Biomonitors have been used for decades to estimate environmental pollution [11]. Mosses as biomonitors have mainly been investigated in their natural environments (passive biomonitoring, e.g. [12]). Nevertheless, active biomonitoring, using moss as transplants, was used in an increasing number during the last years [13–16]. In most of these studies, moss-bags provided satisfying results. Moss-bags were hardly used in environments which are not influenced by natural climatic conditions, like indoor environments. In a pilot study, moss frames were exposed as indicators for road traffic emissions in a road traffic tunnel. Data from this study served to explain surprising results and proofed the applicability of moss as biomonitors, even in the harsh environment of a tunnel [17]. Ahead of the study at hand, indoor monitoring with moss was only performed by Al-Radady et al. [18, 19], who successfully exposed a few moss-bags and subsequently analysed several heavy metals. Two more studies attempted same as the one presented in this article [20, 21].

The study at hand was conducted from 2008–2011. It took place in the Spanish city of Girona. An important goal of this study was to identify a proper tool for indoor monitoring to estimate the pollution exposure of participants of the REGICOR study, a population-based prospective cohort study. Results of moss-samplers monitoring in outdoor environments at the participant's home were already published [22, 23]. However, data on indoor monitoring had not been published since.

The aims of the study at hand were: (1) to test the applicability of moss-samplers for long-term monitoring exposed indoor to quantify indoor heavy metal pollution and (2) to compare outdoor and indoor concentrations of paired samples to estimate outdoor/indoor fluxes of environmental pollutants. Given the wide use of NO_2 as a marker of traffic-related urban air pollution, we compared the findings of the moss study with the parallel measurements of NO_2.

Materials and methods

Moss material

Moss [*Pleurozium schreberi* (Willd. Ex Brid) Mitt.] was collected in March 2008 at a background site (background according to Zechmeister et al. [24]) in the Austrian Alps (Großarl-Valley, 1200 m.a.s.l., E 13°13,9101, N 47°16,9404). The moss was cleaned from any litter and adhering macroscopic particles and reduced to its green shoots only. This method refers to a procedure used in international passive monitoring studies (UNECE ICP-Vegetation, [25]). After homogenizing the samples, metal analysis of a subsample was performed to characterize the background pollution level. The remaining moss was packed in individual plastic bags, containing the amount needed for each moss-sampler, and sent to CREAL—now ISGlobal—together with 43 frames according to the standard procedures established by Zechmeister et al. [17]. The samplers for exposure consist of a wooden frame (inner frame 10×10 cm), equipped with a fine polypropylene net (mesh size 0.9×0.9 cm) containing the moss shoots (approx. 10 mg dry-weight) and were exposed horizontally. The system is shown in Fig. 1.

NO_2-samplers

Nitrogen dioxide was monitored with Palmes diffusion tubes [26] according to the method validated by Bush [27]. The monitor consists of an acrylic tube of 7.1 cm with an intern diameter of 1.1 cm. The atmospheric NO_2 diffuses up the tube, where it gets absorbed on the triethylamine (TEA) coated mesh. Their operation is based on the principle of molecular diffusion, with molecules of gas diffusing from a region of high concentration (open end of the sampler) to a region of low concentration (absorbent end of the sampler), establishing a gradient

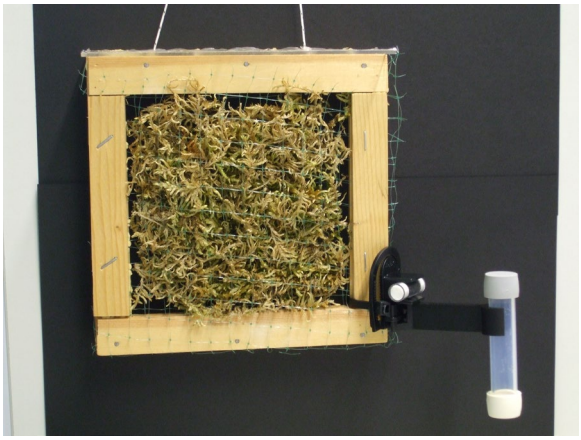

Fig. 1 Wooden frame (10 × 10 cm) containing 5 g of moss; polypropylene net with mesh size 1 × 1 cm; a Palmes diffusion tube is mounted to the frame

(driving force) along the length of the tube. NO_2 diffusion tubes were located next to the moss-samplers, both indoor and outdoor (see Fig. 1). Each tube was exposed for 1 month and then replaced by a new tube deployed during the second month.

Sites of exposure

The geographic area of the study was the city of Girona in northern Catalonia, Spain. The city has a population of about 100.000 and a comparable low Industry. The sampling sites were residences of the REGICOR registry (Girona Heart Registry) participants and research partners [https://regicor.cat/]. Twenty sites were selected according to different levels of air pollution estimated by NO_2 measurements from previous studies and to have a broad distribution across the city (Fig. 2).

Other selection criteria for the sites were: (1) non-smoking household, (2) accessible balcony to deploy outdoor moss, (3) not using gas for cooking, as this may affect indoor pollutants concentration and (4) located on a first or second floor to keep the sampling height constant. To achieve the goal of 20 locations, criteria 3 and 4 were slightly relaxed (e.g. at 25% of sites, gas stoves were situated in a room next to the monitoring room). A series of indoor data were evaluated (e.g. the height of the flat, use of gas stoves, type of windows, type of furniture and floors, habits on aeration and air conditioning, cleaning habits, etc.). However, these data were not available consistently for all flats. Therefore, these data could not be used for statistical analyses, however for the explanation of one or the other surprising result.

Indoor mosses were exposed at convenient sites not interfering with participant's habits in living or sleeping

Fig. 2 Map of the Girona area covered by the moss monitoring; observed sites are marked in yellow; map source: © OpenStreetMap 2020; grid net: WGS 1984 Web Mercator Auxiliary Sphere

rooms. Outdoor mosses were placed at a sheltered location, such as under the roof of the balcony. This placement should avoid any wet deposition (rain) deriving from long-range transport as well as a potential washout of already deposited dry particles. In cases where optimal locations were more exposed to weather, an acrylic roof was fixed on top of the moss frame, using double-sided tape. NO_2-samplers were placed at the moss frame (see Figs. 1 and 3). The exposure period was 8 weeks, between May 5th, 2008 and, July 4th, 2008.

Sample preparation

Moss samples were subjected to microwave digestion with H_2O_2/HNO_3. Sample aliquots of 200 mg plus 4 ml of ultra-pure nitric acid and 1 ml 30% H_2O_2 solution (ultrapure, Merck) were placed into acid-steam-cleaned PFA-microwave digestion vessels and closed with precleaned PTFE-caps. The digestion was performed applying a microwave program employing maximum microwave power of 450 W. After cooling, the digested samples were transferred to PP-vials and filled up to 10 ml with ultrapure water. Microwave digestion blanks were prepared

Fig. 3 Moss-sampler covered by an acrylic roof for outdoor exposure

using ultra-pure water. For quantification multi-element standards were prepared from Merck ICP-Single element standard in PFA bottles and vials through dilution in ultra-pure water and addition of 1% nitric acid. The acid content matched the content in the respective samples. Before ICP-SFMS measurement, indium was added to all samples and standards as an internal standard at a final concentration of 1 µg/l. TM 27.2 (acidified surface water, Environment Canada, National Water Research Institute, Burlington, Canada) was used for calibration quality control.

Chemical analysis

Elemental analysis was carried out on an Element 2 ICP-SFMS (ThermoFisher, Bremen, Germany). As a sample introduction system, a PFA micro-flow nebulizer (Elemental Scientific Inc., Cuming, Omaha, USA) with an i.d. of 45 µm was used during the study. The self-aspirating PFA micro-flow nebulizer (ESI) at a flow of 100 µl/min was combined with a PFA double-pass Scott-type spray chamber, a sapphire injector pipe, a quartz torch, and platinum sampler and skimmer cones (all parts Thermo Fisher). The following ICP-SFMS operating conditions were applied in this study: RF power of 1300 W and plasma gas flow of 16 l/min were applied. Sample gas and auxiliary gas flows were set to 1.06 l/min and 0.86 l/min, respectively. ^{195}Pt at low resolution (LR), ^{27}Al, ^{52}Cr, ^{65}Cu, ^{66}Zn, ^{95}Mo, ^{111}Cd, ^{118}Sn, ^{121}Sb at medium resolution (MR), and ^{75}As at high resolution were selected isotopes for interference-free ICP-SFMS measurement. During all measurements, 115In was used as an internal standard at all resolutions. Nominal mass resolutions of the Element 2 ICP-SFMS for low resolution (LR), medium resolution (MR), and high resolution (HR) are 350, 4500, and 10 000, respectively. Metal analyses were performed at the laboratory of the University of Natural Resources and Applied Life Sciences, Department of Chemistry, Vienna (Austria).

QA/QC: In the measurement sequence about 20% of the samples were related to calibration and quality control. Calibrants, QC standards, blanks, and reference materials were included. 115In was used as an internal standard at all resolutions. Trueness was assessed by measuring the certified reference material TM27.2. The measured concentrations agreed with the certified concentrations within their uncertainty. Due to the internal standardization strategy, intermediate precisions of < 5% for all elements above LOQs could be obtained. Table 1 gives the excellent procedural limits of detection (obtained by replicate bank digestions) proving the method fit for purpose. On average, limits of detection in the low ng/g dry-weight were obtained by sector field ICP-MS. Only the measurement of Al und Zn revealed limits of detection, which were in the µg /g dry-weight range.

Chemical analysis of the NO_2-samplers was performed at the AEA Energy & Environment laboratories in London, UK. The nitrogen dioxide was analysed colorimetrically with the method described by Atkins et al. [28].

Statistical analyses

Principal component analyses (PCA) were performed with not autocorrelated metals and NO_2 with settings for pairwise listing and eigenvalue > 1. The influence of sites on the various components was calculated as well. These tests and their graphical outcomes were performed by using Statgraphics 18. Metals and NO_2, both in indoor and outdoor samples, were correlated by Spearman rank test.

Two types of analyses were performed to compare outdoor with indoor environments at the same sites: (a) the calculation of correlation coefficients between indoor and concentration of the various elements as well as (b) the calculation of the indoor/outdoor ratios which often were taken as fluxes from outdoor to indoor [e.g. [6, 29]. For comparing outdoor and indoor concentrations, a nonparametric test for paired samples (Wilcoxon sign-rank test) was applied. These tests were performed using SPSS Statistics 24.

Levels of significance were stated as $p < 0.001$ (***), $p < 0.01$ (**) and $p < 0.05$ (*).

Table 1 LOD and LOQ for the detection of the various isotopes in moss samples and concentrations of four subsamples (moss A–D) of the moss-material before exposure; concentrations are given in µg/g dry-weight; *LR* low resolution, *MR* medium resolution, *HR* high resolution according the method of analysis

	LOD	LOQ	Moss A	Moss B	Moss C	Moss D
Pt^{195}(LR)	0.0002	0.0007	< LOD	< LOD	< LOD	< LOD
Al^{27}(MR)	1.9460	6.4868	166	132	150	160
Cr^{52}(MR)	0.0075	0.0251	< LOD	< LOD	< LOD	< LOD
Cu^{65}(MR)	0.0108	0.0361	4.45	3.89	4.05	4.33
Zn^{66}(MR)	1.0760	3.5868	12.3	13.7	14.1	14.6
Sn^{118}(MR)	0.0032	0.0107	0.09	0.09	0.09	0.10
Cd^{111}(MR)	0.0182	0.0607	0.04	0.04	0.04	0.04
Pb^{208}(MR)	0.0039	0.0131	2.33	2.41	2.35	2.59
Mo^{98}(MR)	0.0011	0.0037	0.12	0.12	0.10	0.09
Sb^{121}(MR)	0.0032	0.0106	< LOD	< LOD	< LOD	< LOD
As^{75}(HR)	0.0069	0.0230	0.09	0.10	0.12	0.13

Results

Concentrations in moss-samplers

Pre-exposure element concentrations of four subsamples of the moss-material are shown in Table 1. Post-exposure element concentrations of mosses in indoor and corresponding outdoor locations are given in Table 2. Except for Cd, all elements had significantly higher concentrations ($p < 0.01$) in outdoor mosses than at corresponding indoor sites. Pt was below the level of detection (LOD) in most of the cases, therefore, excluded from further evaluation.

Except for As in indoor samples, post-exposure concentrations were higher than in the pre-exposure analyses, in all elements, and both in indoor and outdoor moss samples. For outdoor mosses, the loss of As observed indoors was masked by outdoor deposition. The loss of As during the sampling period might be a result of its anionic form, which cannot be bound permanently by the cationic exchange capacities of the moss. Given the loss of As in moss samples, we excluded this element from further analysis, too.

Mean (SD) concentration NO_2 in outdoor environments was 25.0 µg/m^3 (±9.6) compared to 21.43 µg/m^3 (±6.0) for indoor environments. NO_2 concentrations ranged outdoors from 9.32 µg/m^3 to 44.63 µg/m^3 and indoors from 11.41 µg/m^3 to 36.85 µg/m^3.

Clustering of environmental pollutants

Correlation coefficients between all elements can be seen in Table 3 for outdoor and in Table 4 for indoor measurements. For outdoor environments, most remarkable were the high correlations (all above 0.8***) between Cr and Al, Cr and Sb, Cr and Sn, Sb and Sn, Mo and Sn. For indoor environments, correlation coefficients were high (> 0.8***) for Al and Cr, Sb and Pb, and Cd and Zn. More correlations can be seen in Tables 3 and 4. The only significant correlation of NO_2 with any of the analysed elements was for Sb in outdoor environments (0.554; $p < 0.05$).

In principal component analysis (PCA) for outdoor mosses, component one (50.2%) was mainly driven by Sb and NO_2, whereas component two (26.5%) was driven by Cd (Fig. 4). Sites 495 (component weight 3.9), 22 (2.5), and 164 (1.9) had by far the strongest influence on component 1 and, therefore, were strongly influenced by Sb and NO_2. A different picture was given for indoor mosses. In PCA Sb, Pb and Cr strongly influenced component one (39.7.8%), whereas component two (22.9%) was mainly driven by Mo and NO_2, differentiating from Cu on the negative side (Fig. 5). Sites 496 (3.5), 22 (2.8), 55 (2.7) and 164 (2.5) strongly influenced component 1. Sites 75 (− 2.8), 495 (2.4) and 349 (1.8) were most influential on component 2. As there is only little difference between component weights on axis 1 and 2 the indoor PCA has to be taken with caution.

Comparison of indoor and outdoor concentrations

Correlations between indoor and outdoor concentrations of the different elements in moss at the same site were not significant (Table 5). The highest correlation was observed for Sn (Spearman Rho 0.421). Correlation between indoor and outdoor concentrations of NO_2 in air was not either significant.

The indoor/outdoor ratio can be seen for each element and each home in Table 6. A ratio of < 1 stands for higher outdoor than indoor concentrations in paired samples. On average, outdoor concentrations were slightly higher than indoor concentrations for all elements and NO_2 as well as at each location. In the case of Sn and Sb, this was true at all 20 sites. For all other

Table 2 Concentrations of metals in mosses (µg/g dry-weight) and NO_2 (µg/m^3) in air, as measured by samplers (Fig. 1) exposed for 8 weeks in indoor (_i) and outdoor (_o) environments at corresponding sites at Girona (Spain); site numbers (e.g. 1 iM 017) are according the internal numbers of the REGICOR-study

	Al_i	Al_o	Cr_i	Cr_o	Cu_i	Cu_o	Zn_i	Zn_o	Sn_i	Sn_o	Cd_i	Cd_o	Pb_i	Pb_o	Mo_i	Mo_o	Sb_i	Sb_o	As_i	As_o	NO_2_i	NO_2_o
1 iM 017	332	774	0.51	1.02	6.08	6.99	36.9	33.3	0.18	0.33	0.07	0.08	2.84	3.00	0.21	0.27	0.12	0.21	0.04	0.13	25.6	14.2
1 iM 018	371	392	0.59	0.72	6.30	5.81	29.5	30.1	0.21	0.23	0.06	0.09	2.55	2.65	0.22	0.22	0.12	0.15	0.01	0.07	11.4	18.8
1 iM 021	378	412	0.62	0.67	5.68	6.00	30.6	44.6	0.17	0.25	0.06	0.09	2.22	3.20	0.23	0.23	0.11	0.16	0.06	0.04	26.7	38.1
1 iM 022	614	568	0.77	1.22	7.10	9.56	39.8	40.6	0.22	0.66	0.10	0.08	2.88	3.14	0.22	0.42	0.14	0.48	0.13	0.12	15.6	44.6
1 iM 024	369	639	0.59	0.96	5.96	7.15	31.9	37.7	0.21	0.26	0.07	0.07	2.74	2.62	0.23	0.22	0.12	0.29	0.01	0.55	20.3	38.6
1 iM 027	355	405	0.57	0.65	6.01	5.70	30.0	28.6	0.19	0.21	0.06	0.06	2.45	2.52	0.22	0.23	0.12	0.14	0.01	0.04	23.6	18.8
1 iM 055	472	540	0.75	0.75	6.44	6.99	35.9	32.5	0.23	0.25	0.07	0.06	3.11	2.45	0.24	0.18	0.14	0.15	0.01	0.12	24.4	22.4
1 iM 075	384	464	0.61	0.78	7.73	6.72	36.6	30.7	0.16	0.24	0.06	0.06	2.81	2.62	0.20	0.23	0.13	0.16	0.01	0.04	18.6	18.6
1 iM 113	449	342	0.63	0.66	6.71	6.25	31.4	40.5	0.17	0.25	0.06	0.09	2.03	2.72	0.21	0.38	0.10	0.18	0.04	0.09	22.1	25.7
1 iM 114	404	420	0.62	0.70	6.16	6.57	30.5	30.8	0.17	0.19	0.05	0.05	2.49	2.28	0.17	0.18	0.12	0.13	0.01	0.52	24.5	20.9
1 iM 159	472	626	0.66	0.85	6.44	7.01	28.7	32.6	0.20	0.29	0.06	0.06	2.34	3.18	0.21	0.26	0.12	0.21	0.08	0.11	16.4	23.5
1 iM 164	421	735	0.68	1.86	6.36	11.33	32.4	49.5	0.22	0.80	0.07	0.07	3.15	3.87	0.24	0.44	0.14	0.52	0.04	0.13	28.4	24.8
1 iM 301	345	509	0.54	0.82	5.95	8.24	28.3	32.1	0.20	0.32	0.06	0.05	2.60	2.82	0.23	0.26	0.12	0.24	0.08	0.30	21.9	31.8
1 iM 349	418	381	0.56	0.62	6.32	5.61	38.9	30.0	0.17	0.20	0.10	0.06	2.48	2.62	0.26	0.21	0.11	0.13	0.00	0.08	24.4	28.9
1 iM 350	654	463	0.76	0.73	5.83	6.57	31.7	33.9	0.20	0.27	0.06	0.09	2.49	3.01	0.24	0.26	0.11	0.15	0.02	0.16	12.4	9.3
1 iM 380	410	445	0.62	0.83	5.75	6.07	28.0	28.6	0.18	0.38	0.05	0.07	2.28	3.04	0.23	0.26	0.11	0.20	0.00	0.14	23.2	24.1
1 iM 417	336	505	0.53	0.74	4.88	6.41	34.6	36.9	0.18	0.22	0.08	0.06	2.34	3.20	0.21	0.21	0.11	0.14	0.05	0.22	13.8	15.8
1 iM 419	334	598	0.49	0.84	6.81	7.44	31.5	37.5	0.17	0.28	0.06	0.09	2.62	3.26	0.20	0.25	0.11	0.14	0.04	0.06	15.4	11.5
1 iM 495	486	711	0.68	1.17	5.31	8.99	33.5	41.7	0.21	0.46	0.08	0.09	2.48	6.31	0.23	0.30	0.12	0.34	0.04	0.21	36.9	41.3
1 iM 496	536	494	0.88	0.78	5.83	6.09	37.9	44.7	0.23	0.25	0.09	0.11	2.94	3.16	0.25	0.26	0.14	0.16	0.09	0.13	23.0	27.9
Mean	427	521	0.63	0.87	6.18	7.08	32.9	35.8	0.19	0.32	0.07	0.07	2.59	3.08	0.22	0.26	0.12	0.21	0.04	0.16	21.4	25.0
SD	88	122	0.1	0.28	0.61	1.41	3.542	5.9	0.02	0.15	0.01	0.02	0.29	0.82	0.02	0.07	0.01	0.11	0.03	0.14	6.0	9.6
Min	332	342	0.49	0.62	4.88	5.61	28.0	28.6	0.16	0.19	0.05	0.05	2.03	2.28	0.17	0.18	0.10	0.13	0.00	0.04	11.4	9.3
Max	654	774	0.88	1.86	7.73	11.33	39.8	49.5	0.23	0.80	0.10	0.11	3.15	6.31	0.26	0.44	0.14	0.52	0.13	0.55	36.9	44.6
Median	407	500	0.62	0.78	6.12	6.64	31.8	33.6	0.19	0.25	0.06	0.07	2.52	3.01	0.22	0.26	0.12	0.16	0.04	0.12	22.5	23.8

Table 3 Correlation coefficients of elements and NO_2 at 20 outdoor sites

	Al	Cr	Cu	Zn	Sn	Cd	Pb	Mo	Sb
Cr	0.907***								
Cu	0.845*	0.849***							
Zn	0.452*	0.441	0.484*						
Sn	0.705**	0.858***	0.731**	0.455*					
Cd	0.007	0.122	-0.056	0.578**	0.288				
Pb	0.459*	0.523*	0.387	0.662***	0.616**	0.499*			
Mo	0.382	0.579*	0.482*	0.508*	0.806***	0.446	0.579*		
Sb	0.621**	0.789***	0.673**	0.499*	0.847***	0.191	0.423	0.752***	
NO_2	0.069	0.214	0.209	0.386	0.294	0.062	0.137	0.261	0.552*

Significances given as: $p < 0.001$***, $p < 0.01$**, $p < 0.05$*

Table 4 Correlation coefficients of elements and NO_2 at 20 indoor sites

	Al	Cr	Cu	Zn	Sn	Cd	Pb	Mo	Sb
Cr	0.935***								
Cu	0.102	0.046							
Zn	0.256	0.199	0.206						
Sn	0.449*	0.566*	− 0.113	0.178					
Cd	0.247	0.179	− 0.031	0.834***	0.467*				
Pb	0.092	0.206	0.371	0.539*	0.547*	0.417			
Mo	0.458*	0.421	− 0.349	0.227	0.549*	0.445	0.215		
Sb	0.269	0.431	0.298	0.369	0.658**	0.350	0.806***	0.161	
NO_2	0.054	0.122	− 0.207	0.120	-0.009	0.118	0.036	0.276	0.172

Significances given as: $p < 0.001$***, $p < 0.01$**, $p < 0.05$*

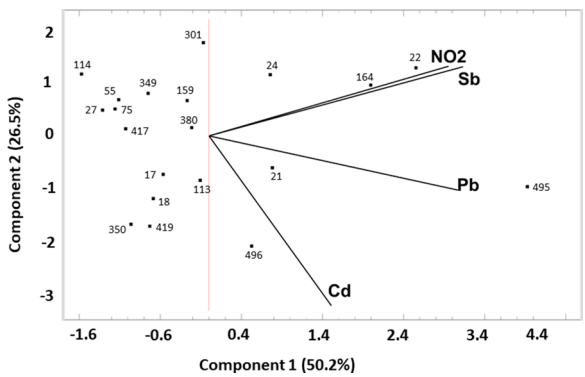

Fig. 4 Principal component analysis of outdoor mosses and sites (autocorrelated elements were excluded); for site numbers refer to Table 2

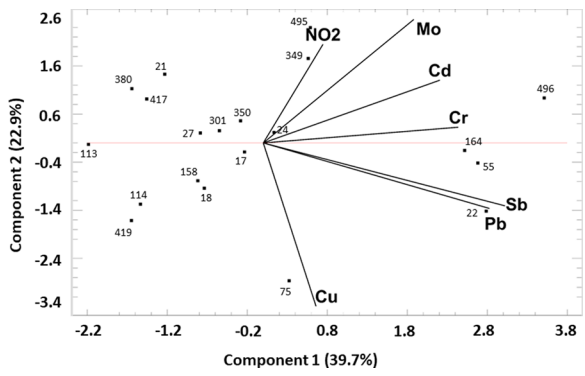

Fig. 5 Principal component analysis of indoor mosses and sites (autocorrelated elements were excluded); for site numbers refer to Table 2

elements, indoor concentrations were higher in, on average, 20.5% of the samples. However, those ratios were mostly close to unity as well. Only in the case of Cd, values varied much more widely (0.64–1.59). The latter value was observed at site iM349 where indeed five of the nine elements showed 10% to 59% higher concentrations indoors as compared to outdoors. Ratios were lowest for Sb (mean 0.66) and Sn (0.69) for outdoor values, occasionally being more than three times higher than indoors. The ratio for NO_2 had a wide variability (0.35–1.81), although the average across sites was close to unity (0.95).

Table 5 Spearman Rho correlation coefficients and corresponding significances for indoor/outdoor correlations for the analysed elements and NO_2 at 20 sites

	Spearman Rho correlation coefficients	Sign
Al	− 0.040	.867
Cr	0.189	.439
Cu	0.257	.274
Zn	0.364	.115
Sn	0.421	.065
Cd	0.168	.480
Pb	− 0.078	.744
Mo	0.077	.748
Sb	0.389	.090
NO_2	0.373	.105

Discussion

Outdoor situations

Comparing initial concentrations before exposure with concentrations after exposure (Tables 1 and 2), elevated loads of metals were found in all outdoor mosses. There were significant correlations between Cr, Mo, Sb, and Sn in moss and traffic densities based on the data set at hand, evaluated by Rivera et al. [22, 23]. There were significant correlations between these metals and average daily traffic (r: 0.81, $p < 0.0001$) and the bus lines circulating in the nearest street (r: 0.91, $p < 0.0001$]) [23]. Sites with high traffic densities reflect this scheme as well (see Fig. 4). It can be concluded that at least Sb, Sn, Cr, and Mo reflect emissions from road traffic, a finding confirmed in other studies from urban environments [17, 30, 31].

Nitrogen dioxide concentrations have been widely used as indicators for traffic-related air pollution [32, 33]. There were significant correlations between NO_2 and Sb concentrations in our outdoor moss samples (0.554, $p < 0.05$). With outdoor NO_2 being dominated by traffic in the city of Girona, our data confirm the use of Sb as another marker of local traffic-related pollutants.

In Table 7, concentrations in moss-samplers of equivalent exposure periods as reported in other studies are compared with our results. This table should be interpreted with caution as absolute contents of metals in transplanted mosses may not be comparable in different studies because initial contents could be different. Nevertheless, it gives an approximative overview of pollution loads in various cities. Concentrations found at the Girona site were comparably lower than in other cities shown in this table. This discrepancy is probably caused by lower pollution levels in the rather small city of Girona (e.g. low traffic burdens), than in the mostly larger cities compared. Additionally, moss-samplers were exposed to different heights in the listed cities. Lacic et al. [34], Capozzi et al. [15], or Goryainova [35] found a significant decrease of exposure with height in urban street canyons. Our exposure sites had an average height of 8.7 m (ranging between 3.7 and 20 m) above street level, compared to mostly < 4 m above ground level in the other studies, which is another reason for lower pollution levels in Girona measured with moss-samples.

Indoor versus outdoor concentrations

Different elements showed different concentrations in indoor and outdoor environments (see Table 5). Overall, outdoor contents were much stronger intercorrelated than indoor contents (Tables 3, 4). Thus, outdoor sources of pollution determined more strongly the metal contents in moss than indoor sources. The scarcity correlations between metal contents indoors may suggest that indoor sources were more diffuse, although more or less identifiable.

In theory, in the absence of indoor sources and of any sealing of the indoor environment from outdoors, one would expect highly correlated indoor/outdoor pairs and the same concentrations for all indicators, thus, indoor/outdoor ratios at unity, with indoor values, thus, explained by outdoor air pollution [34]. Higher indoor concentrations—coupled with poor indoor/outdoor correlations would point toward indoor sources of those pollutants.

Sb (mean 0.66) and Sn (0.69) showed the lowest i/o ratios. This ratio suggests that, for these elements, outdoor sources of pollution were much more important than indoor sources. Both elements derive mainly from traffic emissions, especially abrasions of brake-wear (Sb) and car-bodies (Sn) [17, 36, 37]. Traffic is also a source for Cr and Mo, whose outdoor contents were intercorrelated (Table 3). Compared to urban sites in Capozzi et al. [21], one of the very few corresponding studies, the ratio for Cr is lower in their study. This low ratio suggests an even stronger contribution from

Table 6 Quotient of indoor/outdoor concentrations (I/O) per element and home; quotients for NO_2 are also shown; site numbers (e.g. 1 iM 017) are according the internal numbers of the REGICOR-study; %—% of sites with higher indoor values

	Al	Cr	Cu	Zn	Sn	Cd	Pb	Mo	Sb	NO_2	I/O per home
1 iM 017	0.43	0.50	0.87	1.11	0.55	0.94	0.95	0.76	0.57	1.81	0.70
1 iM 018	0.94	0.82	1.08	0.98	0.91	0.64	0.96	0.97	0.81	0.61	0.83
1 iM 021	0.92	0.92	0.95	0.69	0.70	0.72	0.69	1.00	0.69	0.70	0.88
1 iM 022	1.08	0.63	0.74	0.98	0.34	1.13	0.92	0.53	0.28	0.35	0.77
1 iM 024	0.58	0.61	0.83	0.85	0.82	0.91	1.05	1.01	0.41	0.53	0.71
1 iM 027	0.88	0.88	1.05	1.05	0.89	1.01	0.97	0.94	0.87	1.25	0.87
1 iM 055	0.88	0.99	0.92	1.10	0.94	1.24	1.27	1.33	0.95	1.09	0.97
1 iM 075	0.83	0.78	1.15	1.19	0.69	1.11	1.07	0.86	0.85	1.00	0.89
1 iM 113	1.31	0.95	1.07	0.77	0.66	0.68	0.75	0.56	0.58	0.86	0.77
1 iM 114	0.96	0.88	0.94	0.99	0.87	0.95	1.09	0.97	0.89	1.17	0.86
1 iM 159	0.75	0.78	0.92	0.88	0.70	1.04	0.74	0.81	0.58	0.70	0.79
1 iM 164	0.57	0.37	0.56	0.66	0.28	1.10	0.82	0.55	0.26	1.15	0.55
1 iM 301	0.68	0.66	0.72	0.88	0.61	1.16	0.92	0.86	0.51	0.69	0.73
1 iM 349	1.10	0.90	1.13	1.30	0.88	1.59	0.95	1.24	0.83	0.84	1.00
1 iM 350	1.41	1.04	0.89	0.94	0.76	0.69	0.83	0.94	0.76	1.33	0.84
1 iM 380	0.92	0.75	0.95	0.98	0.49	0.82	0.75	0.87	0.55	0.96	0.71
1 iM 417	0.67	0.72	0.76	0.94	0.82	1.37	0.73	1.02	0.81	0.87	0.80
1 iM 419	0.56	0.59	0.92	0.84	0.60	0.68	0.80	0.80	0.77	1.34	0.72
1 iM 495	0.68	0.58	0.59	0.80	0.45	0.88	0.39	0.76	0.36	0.89	0.57
1 iM 496	1.09	1.13	0.96	0.85	0.94	0.82	0.93	0.96	0.87	0.82	0.92
Average I/O	0.86	0.77	0.90	0.94	0.69	0.97	0.88	0.89	0.66	0.95	0.79
%	25	10	25	25	0	45	20	25	0	40	

Table 7 Comparison of concentrations (µg/g dry-weight) in moss bags and moss-samplers (Girona) after approx. 2 months exposure at various cities; [1]—[35], [2]—[47]. [3]—[48], [4]—[37], [5]—[49], [6]—[50]; *Nr* number of sites in the respective study

	Nr	Al	Cr	Cu	Zn	Cd	Pb	Mo	Sb	As
Girona (this study)	20	653	1.11	8.27	37	0.08	3.3		0.31	0.17
Moscow[1]	3	970	2.5	5.2	50.3	0.1	4.6	0.2	0.4	0.6
Turku[2]	35	765	5.7	14.4	61.7		1.7			
Belgrade[3]	3	1040	2.6		63				2.6	0.38
Belgrade[4]	48		2.6	15.7	210		6.5		1.8	0.3
Naples[5]	22	2922	4.4	30	157	0.7	76	2.4		
Trieste/Naples[6]	2	876	5.03	31.3	104	0.3	16.9			

outdoor sources than in our study or an influence of measurement error in our small study. They state traffic as the main driver for Cr, too. In studies relying on technical measurements, traffic burdens show I/O ratios below one [29]. This fact is underpinned by a study of Chen & Zhao [9], who show lower I/O values for fine particles than coarse ones—traffic derived pollutants are often associated with (ultra)fine particles.

Other elements (Cd, Zn) and NO_2 showed ratios closer to the unit. These cases suggest equilibrium between outdoor and indoor concentrations of atmospheric pollutants (Table 6). Cd values (mean ratio 0.97) ranged from a low ratio of 0.64 to a high one of 1.59, and indoor/outdoor correlations were very low. High indoor values were observed at site iM349, where five of the nine elements showed 10% to 59% higher values

indoors as compared to outdoors, pointing toward some relevant indoor source. For NO_2 (mean ratio 0.95), the site-specific values ranged from 0.35 to 1.81, coupled with a modest correlation of 0.373. Measurement precision of NO_2-samplers is high, and penetration from outdoor to indoor typically high, thus as shown in other studies [38], in the absence of indoor sources, i/o correlations tend to be higher. Given the limited sample with only 20 sites, the study focused on sites with supposedly no indoor sources. Preselection criteria of monitoring sites included no smoking homes and predominantly no gas stoves or open fires at the exposure rooms. Results indicate that some sites were indeed influenced by indoor NO_2 sources such as gas cooking and smoking. Unfortunately, the study could not include time-activity/exposure diaries to evaluate further the contribution of these indoor sources. These sources may also explain the occasionally higher concentrations of elements on mosses. Thus, as shown by others, the value of indoor NO_2 as a marker of outdoor (thus traffic-related) NO_2 concentrations largely depends on the absence of indoor sources of NO_2 [38, 39].

There were particular cases (polluted homes) and technical reasons (a small number of samples, only one gaseous pollutant measured) disturbing the general pattern described above. Our study underscores on one side the important role of outdoor pollution as a determinant of the indoor conditions and on the other side the potential relevance of individual indoor sources. Measurement errors in moss analysis, caused by analytical criteria, can be excluded for most metals. There is a long tradition in quality assurance (e.g. [40, 41]), and instrumental quality was very high, especially in this study. It is more likely that varying indoor exposure conditions (e.g. microclimate) lead to an unbalanced uptake of elements, and capturing of particles differed slightly under these conditions. These causes were investigated and discussed intensively in previous articles (e.g. [42, 43]).

Conclusions and consequences for future indoor monitoring

The study at hand showed that moss exposed at indoor sites could be a promising tool for long-time biomonitoring. However, it had also identified some drawbacks that should be taken into account in future indoor studies.

The long-term exposure is a huge advantage as compared to technical tools with demanding infrastructure and maintenance usually applicable in short-time assessments only. A drawback of the moss method is the relatively small increments of pollutants compared to the pre-exposure concentrations for several elements, at least in the rather clean environments seen nowadays in Western countries. Moss used for monitoring purposes comes from background areas, however sites classified as "background sites" are hardly ever without pollution [38] and increasingly similar to urban areas. Therefore, increments on pre-exposure concentrations can only be observed after long exposure periods or at high pollution levels. Moreover, pollutant concentrations in indoor environments are mostly low, and accumulations can often hardly be seen even after a 2-month exposure period. This low accumulation goes for a series of elements like As or Pt in our study. The EU-FP7 project "moss clone" provided such "clean" material by cloning a moss species [44–46] and tested several methods for optimal exposure [15, 16]. This cloned material should be a proper material for indoor monitoring, however it has never been tested for this purpose yet. Very long exposure periods do not solve the problem of small increments. As shown by Capozzi et al. [15], the best exposure period for active monitoring is 3 months and could be the best option for indoor monitoring too.

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Authors' contributions
HGZ: data evaluation, writing of the manuscript, supervisor for moss affairs. MR: data acquisition during the overall study, analysis of data, contributions to the text. GK: analyses of the moss samples, contribution to the text. JM: contribution in the set-up of the study, contribution to the text. NK: project coordinator, substantial contributions to the conception of the study, contribution to the text. All authors read and approved the final manuscript.

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Availability of data and materials
The datasets generated and analysed during the current study is in major parts included in the presented study, some data are not publicly available due to violating the privacy of participants of the cohort study.

Ethics approval and consent to participate
The ethics committee study Fundació la Marató de TV3 approved the study on September 4th 2008; approval see also attachment.

Consent for publication
Not applicable.

Competing interests
The authors declare that they have no competing interests.

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