GeoHealth

Key Points:

- Magnetic methods and carbon analyses evidence high concentrations of indoor and outdoor PM1 due to traffic sources
- Proximity of traffic source and absence of ventilation favorize accumulation of PM indoor threatening children's health and learning skills
- Passive monitoring using vegetal media facilitates teacher and pupil involvement and prevents eco-anxiety

Barking up the Right Tree: Using Tree Bark to Track Airborne Particles in School Environment and Link Science to Society

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Abstract Children's exposure to air pollution affects both their health and learning skills. Fine and ultrafine particulate matter (PM2.5, PM1), notably issued from traffic sources in urban centers, belong to the most potential harmful health hazards. However their monitoring and the society's awareness on their dangers need to be consolidated. In this study, raising teacher and pupil involvement for air quality improvement in their schools environment is reached through developing a passive monitoring technique (bio-sensors made of tree bark). The experiment was implemented in two urban elementary schools situated close to a main traffic road of the city of Toulouse (South of France). Magnetic properties, carbonaceous fraction measurements, and scanning electronic microscopy (SEM-EDX) investigations were realized both on passive bio-sensors and filters issued from active sampling. We find that traffic is the main PM1 source for both outdoors and indoors at schools. Higher levels of outdoor PM in the school's environments compared to urban background are reached especially in the cold period. The schools proximity to a main traffic source and lack of ventilation are the main causes for observed PM1 accumulation in classrooms. The co-working experiment with educational teams and pupils shows that the use of bio-sensors is a driver for children empowerment to air pollution and therefore represents a potential key tool for the teachers though limiting eco-anxiety. As PM accumulation is observed in many scholar environments across Europe, the proposed methodology is a step toward a better assessment of PM impact on pupil's health and learning skills.

Plain Language Summary Children's exposure to air pollution affects both their health and learning skills. Monitoring airborne particles in school environments from a knowledge co-production perspective seems essential to address air pollution in schools. Here we use tree bark and filter samples to monitor air quality in schools. Teachers and pupils are involved in the implementation process. The experiment was set up in two schools in Toulouse (France), close to the busy ring road. Traffic is the main source of air pollution both inside and outside of the classrooms. Quantities of airborne particles are higher during cold periods. The proximity of the highway and poor ventilation in the schools are the main cause of airborne particles accumulation inside classrooms. Collaboration with school staff and students shows that the use of tree has been a driving force for children to embrace the project and understand the context of air pollution, limiting anxiety related to this topic.

1. Introduction

Children are particularly vulnerable to air pollution exposure (Ferguson & Solo-Gabriele, 2016; Kelly & Fussell, 2015; Khreis et al., 2017; Szabados et al., 2021), one of the most important environmental causes of premature death in Europe (European Environmental Agency, 2021). They have higher air intake in comparison to adults, leading to higher absorption of pollutants through respiration, besides having immature respiratory and neurological systems (Kim et al., 2004; Martins et al., 2020; Rice & Barone, 2000). Health consequences in infants of exposure to pollutants and particulate matter (PM) may include respiratory diseases, impairments on their cardiovascular system and worsening of their neuropsychological development (WHO, 2021b and references therein). In terms of health hazards, the fine and ultrafine fraction are the most harmful ones (Martins et al., 2020; Pope & Dockery, 2006) as they may penetrate deeper into the respiratory system. The translocation from the

lungs to the bloodstream is more likely to happen in poorly soluble ultrafine particles (Pope & Dockery, 2006), thereby enhancing the deleterious effects of associated hazardous substances.

Ensuring good air quality for children's environments also provides optimal conditions for learning (Annesi-Maesano et al., 2013; Höfner & Schütze, 2021; Mendell & Heath, 2005; Vornanen-Winqvist et al., 2020), in addition to preserving their health. However, PM2.5 concentrations (i.e., particle sizes smaller than 2.5 μm) inside classrooms could be higher than outdoors, at least in European schools (Kalimeri et al., 2019). Children spend a lot of their school time indoors, making their indoor exposure to PM as important as the outdoors one. In France, students between 4 and 14 years old spend on average 3.8 hr of their daily time in schools (Zmirou et al., 2002). For a non-negligible part of the children, it is common that this time represents 10 hr on a weekday (as it includes childcare in schools). It is therefore important to assess the level of fine and ultrafine particles (PM2.5 and PM1) in air inside and outside classrooms as well as identify practices that can improve air quality.

Children's exposure depends on local outdoor air quality, indoor sources, and the ventilation system (Dimitroulopoulou, 2012). Children's lung can notably be irreversibly impacted by local traffic exposure, regardless of regional air quality (Gauderman et al., 2007). Manual ventilation of the classroom by opening the windows for example, can also induce inconveniences (temperature, wind, and ambient noise outside), which can affect the pupils' learning conditions and academic performance (Mumovic et al., 2009). Raising awareness about contaminants, sensors and the role of ventilation is not enough to create leverages toward a more active approach (Tham, 2016). Direct contextualization has been shown to be important (West et al., 2020) in environmental education. Environment education also needs to account for the risk of eco-anxiety raise (Pihkala, 2020). In the light of these facts, in this study, the approach chosen to study air pollution was to use vegetation media as bio-sensors or bioindicators (Chaparro et al., 2020; Nakazato et al., 2018; Parviainen et al., 2020). Environmental magnetism methods can be applied to natural media. They have been used to monitor anthropogenic PM trapped on vegetation such as leaves, lichens and bark (Chaparro et al., 2020; Dawai et al., 2021; Limo et al., 2018; Moreno et al., 2003; Muñoz et al., 2017; Sagnotti et al., 2009; Urbat et al., 2004; Vezzola et al., 2017; Zhang et al., 2008). Such methods rely on tracking magnetic minerals, for example, iron oxides like magnetite, hematite and maghemite (Leite et al., 2018; Marié et al., 2018; Winkler et al., 2021). For traffic-related sources, these magnetic minerals can originate from internal fuel combustion of vehicles, disk brakes abrasion and erosion of the pavement.

This study was carried out on two primary schools in Toulouse using both environmental magnetism, carbonaceous fraction measurements and scanning electronic microscopy (SEM-EDX) investigation. Our aim is to explore how the use of environmental magnetism using bio-sensors (tree bark) in addition to conventional techniques helps in transformative ways to tackle air pollution in schools by assessing the indoor/outdoor transfer of particles from traffic and by co-working with teachers and children.

2. Materials and Methods

2.1. Location, Meteorological, and Pollutants Conditions

The city of Toulouse and its surroundings stands on quaternary alluvial terraces incised by the Garonne river. The terrace substratum is made of molasse consisting in fluvial deposits made of clay, marls lacustrine/palustrine limestones and channelized sandstones and conglomerates derived from the North Pyrenean foreland basin (Christophoul et al., 2014 and references therein).

Toulouse is the fourth biggest city in France, with a metropolitan population of approximately 1,331,000. The ring road constitutes one of the main traffic roads of Toulouse with a total of 35 km, linking important state-highways to the city traffic. It has three lanes in each way, with a mean flow of ca. 120,000 vehicles per day (in 2019) on its southern part (https://www.aua-toulouse.org/wp-content/uploads/2022/02/AUAT_chiffres_transports_2022.pdf, consulted on January of 2022).

The two schools, separated by 1.3 km, are located south of downtown Toulouse, on the east bank of the Garonne river. School 1 and School 2 are located approximately 150 and 300 m north and northwest of the Toulouse ring road, respectively. Both schools' courtyards are planted with Platanus trees (Figure 1). In School 1, the total number of pupils is around 360, divided into 15 classes with an average of 24 pupils per class. This school has two courtyards, separated by a school building. The West and the East courtyards have an area of 1,400 and 3,800 m^2,
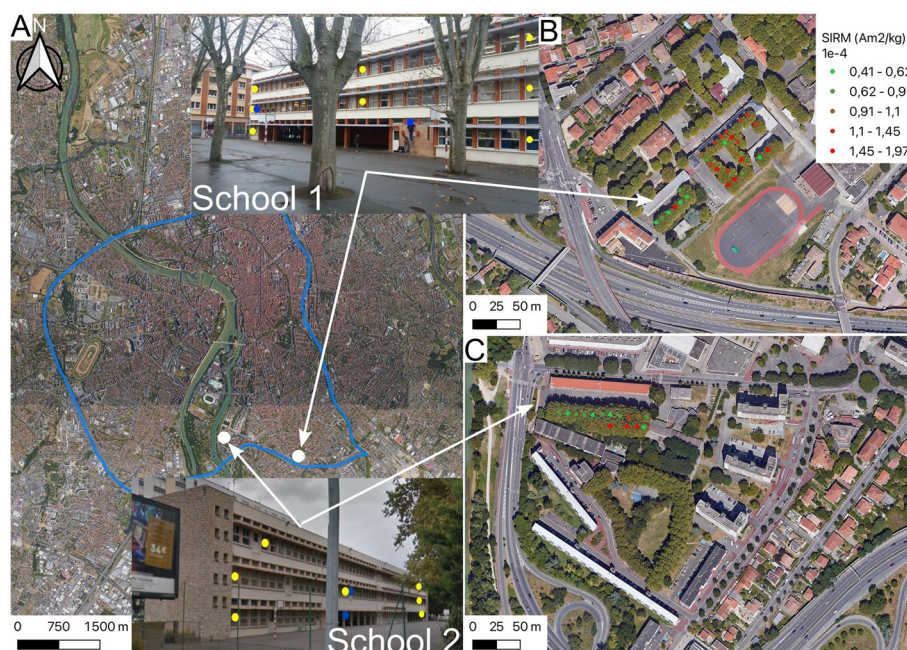
Figure 1. (a) Satellite view of Toulouse (modified from Google Earth) with school locations and photographs of instrumented school buildings (insets). The ring road is highlighted in blue. Position of bio-sensors (yellow) and PM1 air filters location (blue) are marked with dots on the pictures. (b and c) Zoom on Schools 1 and 2 locations. $SIRM_M$ values obtained on tree barks from the sampled courtyard trees are displayed. The same color scale is used for schools 1 and 2.

respectively. School 2 counts 140 pupils for 8 classes, with classrooms of 12–26 students. It has one courtyard with an area of 4,400 m^2. School buildings from School 2 and bordering West courtyard of School 1 were built in the 1950s in the same architectural-style and materials. They have classrooms of around 58 m^2. Buildings from East courtyard of School 1 are historical buildings from the 1930s. In this study, courtyards and selected classrooms were sampled with bio-sensors, tree bark and air filters. In School 1, the bio-sensors and air filters were placed only in the school building bordering the West courtyard.

Concerning meteorological conditions, Toulouse has a continental climate and is located in a ventilation corridor sometimes affected by either SE wind from the Mediterranean Sea or NW winds from the Atlantic Ocean. Winters are cold and cloudy with degraded air quality due to low ventilation and emissions increase from traffic and domestic heating (https://www.atmo-occitanie.org). During spring and early summer, air quality improves due to increased ventilation and rainfall. Experiments were launched between the end of 2018 and the first half of 2019. Meteorological parameters from Blagnac Airport and air quality stations from ATMO OCCITANIE in Toulouse (Figures S1 and S2 in Supporting Information S1) show the differences between the sampled periods for NO$_2$, PM10 and PM2.5 concentrations, daily rainfall and wind speed.

2.2. Methodology

Both active and passive sampling methods were deployed. The active sampling consisted in the deployment of Cyclone air filtration pumps with a cutoff size of PM1. Two different sets of samples were used for the passive sampling. The first set was composed of tree collected from the Platanus trees planted in the school courtyards, and the second one consists of 15 couples of bio-sensors made of non-contaminated Platanus tree bark squares developed in the framework of the NanoEnvi project (https://nanoenvi.omp.eu). Using both Platanus tree bark and bio-collectors derived from the co-working experiment with educational teams and pupils.

Low volume (2 L.min^{-1}) Cyclone (BGI by Mesa Labs) with a cutoff size of 1 μm in diameter (PM1) and air filtration pumps (SKC Air Touch) were used for the active sampling. Two sampling lines were run in parallel, the first one equipped with a Whatman quartz filter and the second one with a Teflon (Polytetrafluoroethylene or PTFE filter). The lines were continuously active over one or two weeks, in January/February (period 1) and

April/June (period 2) of 2019, with six filtration lines in each season. In each school, they were deployed in one classroom hosting between 20 and 25 children (indoor, at around 2 m high) and outdoor in the courtyard (at a height of approximately 2.5 m; Figure 1). PM1 mass deposits were measured with the gravimetric method, with a high precision scale (SARTORIUS MC21S). The filters were kept in the weighing room for 24 hr at an ambient relative humidity of $30 \pm 15\%$ before the mass measurement. Atmospheric concentrations are estimated by dividing the mass deposit by the sampled volume. Concentrations of the carbonaceous fractions were achieved through optico-thermal analyses (IMPROVE protocol allowing the separation of the carbonaceous components, Chiappini et al., 2014; Ouafo-Leumbe et al., 2018) with a DRI model 2001 thermal/optical carbon analyzer (Atmoslytic Inc., Calabasas, CA) (Chow et al., 1993, 1994, 2004, 2006). The DRI analyzer has a detection limit of 0.4, 0.1 and 0.3 $\mu g C.cm^{-2}$ for Total Carbon (TC), Elementary Carbon (EC) and Organic Carbon (OC), estimated through the instrument blank. Our measurements have an estimated accuracy of 5% for TC and 10% for EC and OC.

The capacity of bark to accumulate the airborne particles was already demonstrated for Neem trees by Dawaï et al. (2021) in Cameroon, and for various street trees by Chaprarro et al. (2020) in Buenos Aires (Argentina) and in Milan (Italy) by Vezzola et al. (2017). Here, we demonstrated that bark also efficiently accumulates PM in indoor environments as reflected by saturation isothermal remanent magnetization (SIRM) values and SEM images.

Courtyard tree barks were collected for magnetic measurements on each available tree in November and December of 2018 for School 2 and School 1, respectively. The trees are solely from the species Platanus × acerifolia, whose bark has an annual growth cycle. A total of 48 trees were sampled (32 in School 1 and 16 in School 2). In School 1, 7 trees were sampled in the West courtyard and 25 trees in the East courtyard. In School 2, the trees sampled form two parallel lines. School 1 trees have different arrangements on each courtyard. The West one displays a single line parallel to the building, and the East one presents trees on a square disposition around the courtyard (Figure 1). Around 30 g of tree bark per tree was collected between 1 and 2 m above the ground. A small amount of each bark sample was pulverized into a fine homogeneous powder, and then put in two different gel caps sizes, with a mass of 0.1–0.8 g.

The bio-sensors consist of garlands composed of $5/6 \sim 4$ cm^2 squares of Platanus × acerifolia species bark pieces that are suspended from a nylon thread. The tree bark used to build the bio-sensors was collected in areas far from traffic perturbations and had SIRM measured under 1T fields to assess their low level of magnetic content. Bio-sensors were exposed in December 2018 and July 2019 (7 months), in a total of 8 rooms in School 1 and 7 rooms in School 2 (Figure 1). For each classroom, two bio-sensors were placed, indoors and outdoors, at heights of approximately 2 m to prevent children from touching them. After collection, they were ground into a fine, homogeneous powder. The powders were put into gelcaps, with masses ranging from 0.8 to 0.1 g in each gelcap.

Most of the measurements were realized at the magnetic platform at GET (University of Toulouse). Complementary analyses were performed at the paleomagnetic Laboratory at the USP (Sao Paulo, Brazil). SIRM was acquired on filters and courtyard tree bark using both a JR-6 and a JR-5 spinning magnetometer, with 2.4 μA/m sensitivity. For the JR-6 measurements, the high sensibility mode was used, and each measurement was repeated five times. A pulse magnetizer (Magnetic Measurements Ltd.) was used to induce the SIRM, with a saturating magnetic field of 1T. The SIRM gives a qualitative concentration of the magnetic carriers (Evans & Heller, 2003). PTFE filters were chosen to carry out the magnetic investigation since in a pilot conducted in blank filters the PTFE filters had, on average, 20% the SIRM of quartz filters after 1T field induction. Filters were normalized by pumped air volume in the filters (SIRM$_V$). Courtyard tree bark was mass normalized by sample mass (SIRM$_M$).

Hysteresis cycles and back-field remanence curves were measured with a Vibrating Sample Magnetometer (μ-VSM) 3900 from Princeton Measurements Corporation at the IPGP-IMPMC Mineral Magnetism Analytical Facility (Paris, France). The μ-VSM has a sensitivity of 0.5×10^{-9}Am^2. Hysteresis parameters for tree bark and bio-sensors were then calculated, namely the saturation magnetization (M$_S$) and coercive force (H$_C$). The remanent saturation magnetization (M$_{RS}$) and coercive remanent force (H$_{CR}$) were obtained from the backfield remanence curve. The ratio between the hysteresis parameters (M$_{RS}$/M$_S$/and H$_{CR}$/H$_C$) when plotted against each other creates the so-called Day Plot (Dunlop, 2002).

Bio-sensors magnetic measurement protocol consisted of alternating field (AF) demagnetization with 100 mT peak field, followed by a one-step anhysteretic remanent magnetization (ARM) acquisition and then isothermal

remanent magnetization (IRM) acquisition. The measurements were done in a 755-1.65 DC SQUID magnetometer (2G enterprises), with a sensitivity of $10^{-12} Am^2$, located in a magnetically shielded room with an ambient field inferior to 500 nT. The ARM acquisition was performed with a superimposed AF field of 100 mT under a bias field of 30 µT. The ARM magnetization when divided by the bias field gives the susceptibility of the ARM, xARM (Evans & Heller, 2003). The IRM inductions were imparted on a pulse magnetizer (Magnetic Measurements Ltd.). All measurements were done three times and then averaged. The samples were induced in the pulse magnetizer at room temperature, for the acquisition of IRM, with field values of 1T for the SIRM and 300 mT for the backfield IRM (IRM_{-300mT}).

The S-ratio gives the relative contribution of high and low coercivity magnetic minerals to the magnetization. The S-ratio is given by IRM_{-300mT}/SIRM, as at 300 mT only soft coercivity magnetic carriers (magnetite, for instance) will align their magnetic moments with the applied magnetic field, and at 1T all magnetic minerals (soft and hard coercivity carriers) will align their moments with this field (King & Channell, 1991). The xARM/SIRM ratio is widely used as a proxy for grain size variation on magnetite-like magnetic minerals (Maher, 1988) because of its ability to distinguish between coarse and fine grain sizes.

Surface characteristics of the carbonaceous aerosols and of the iron oxides were obtained from representative samples of PM1 filters and bio-sensors through SEM observations using a Field-emission gun FEG SEM JEOL JSM 7100F at the Microcaracterization Center Raimond Castaing (https://centre-castaing.cnrs.fr). Composition characterization was obtained through energy dispersive X-ray spectrometer (EDS) measurements using an Oxford Instrument Detector ($X_{Max} = 80 \ mm^2$ and Ultim $_{Max} = 100 \ mm^2$). The samples were fixed using silver paint and coated with carbon.

2.3. Involvement of Pupils and Teachers in the Experiments in Schools and Social Assessment

The rollout of the experiment to the schools was conducted in several phases. After contacting the city councilor responsible for the elementary school, the first step was to establish a relationship with the school principals via the parent association (School 1) and via the local community association (School 2). The second step consisted in the visit of researchers into the school. This intervention, aiming at presenting the project to pupils and teachers, was carried out for classes of children 9–12 years old (6 classes for School 1 and 5 classes for School 2). The intervention was designed as follows: a first part (20 min) of presentation on the air quality, airborne particles and the project, a second part composed of experimentation workshops and scientific manipulations and an artistic approach, concluded in the third part by general discussion. The workshops were designed to provide an understanding of the project process by discovering the magnetic phenomena (different games and experiments with magnets and ferrofluids), the measurements by the students on a Bartington MS2 field magnetic susceptibility meter of bark samples localized on a map and by extracting magnetic particles from mud. The installation of the active pumps was the subject of a short lecture and discussion in the selected school class of each school during a third intervention. The final step involves the reporting of the results. This was only done in the two classes that hosted the pumps, as the COVID health crisis blocked possible external interventions during this time.

Teachers from the two classes that hosted the pumps were interviewed in a semi-structured manner to assess their awareness of air quality, the ventilation habits they implemented in the classrooms, and the impact of the experiments on the children and teachers.

3. Results

3.1. Air Filters

Outdoor PM1 concentration from period 1 (January–February) ranges from 41.1 to 49.2 µg.m^{-3}, while outdoor PM1 concentration from period 2 (April–June) ranges from 21.1 to 49.8 µg.m^{-3} (Table 1). Indoors, concentration values reveal higher levels during period 1 (varying from 35.7 to 40.7 µg.m^{-3} for School 2 and 1, respectively) than during period 2 (ranging from 24.2 to 29.3 µg.m^{-3} for School 2 and 1, respectively). During the first half of the year 2019, the ambient daily mean concentrations of NO_2 and PM2.5 recorded by AtmoOccitanie at the traffic station (route d'Albi—about 8 km to the North of the schools) range between 8.0 and 80 µg.m^{-3} and 2.0 and 44 µg.m^{-3}, respectively (Figure 2). This rather large daily variability reflects the proximity of traffic emissions.

Table 1
Elemental (EC) and Organic Carbon (OC), PM1 Concentrations, and SIRM Measured at School #1 and #2 on PTFE Filters During the 4 Field Experiments in 2019

Date		EC (μgm^{-3})	OC (μgm^{-3})	OC/EC	PM1 (μgm^{-3})	SIRM (10^{-10} Am^2)
School #1						
14–26 January	Indoor	0.7	4.7	6.7	40.7	2.87
	Outdoor	1.6	4.4	2.7	49.2	3.21
	I/O (no unit)	0.4	1.1	–	0.8	0.9
08–18 April	Indoor	0.8	5.5	6.9	29.2	0.97
	Outdoor	0.9	2.9	3.2	21.1	0.49
	I/O (no unit)	0.9	1.9	–	1.4	1.9
School #2						
28 January–09 February	Indoor	0.4	4.9	12.2	35.7	1.08
	Outdoor	1.4	3.1	2.1	41.1	1.98
	I/O (no unit)	0.3	1.6	–	0.9	0.5
17–27 June	Indoor	0.7	2.8	4.0	24.3	0.42
	Outdoor	0.8	2.6	3.2	49.8	1.94
	I/O (no unit)	0.9	1.1	–	0.5	0.2

Note. The ratio between indoor and outdoor concentration (I/O) is indicated for each parameter and sampling period in 2019.

The average PM2.5 concentration during the sampling periods of the filters (see Table 1) is on average 3 times lower than the PM1 filter concentration.

Figure 2 presents the measured PM1 concentration on the school filters alongside the daily measured PM2.5 and NO$_2$ concentration at official air quality stations (ATMO OCCITANIE). The station is located in Toulouse (route Albi) and represents the traffic background of the city. Generally, the school filters display higher PM1 concentrations during period 1 compared to the PM2.5 measurements from the air quality stations. During period 2, the PM1 concentration on the school filters is comparable to the highest PM2.5 concentration values from the air quality stations. This shows an overall higher concentration of PM in the school environments compared to the urban background of Toulouse. The NO$_2$ concentration patterns follow the PM1 concentration patterns measured in both schools.

The elemental carbon (EC) indoor concentrations reach a maximum value (0.8 μg.m^{-3}) in School 1 during period 2. The minimum concentration is reached in School 2 during period 1 (0.4 μg.m^{-3}). In School 1, the values are
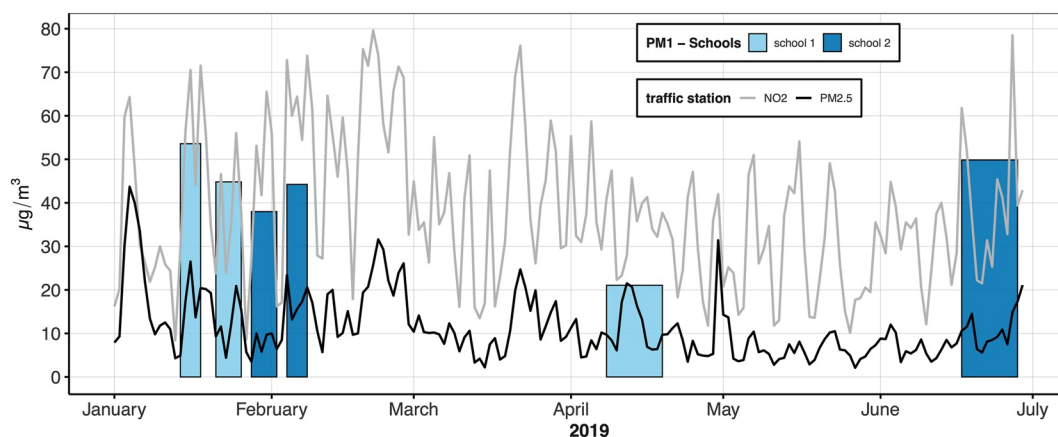

Figure 2. Daily PM2.5 (black line) and NO$_2$ (gray line) concentrations measured at a traffic station in Toulouse (route Albi). Weekly PM1 measured at the schools between January and July 2019 on filters (blue vertical bars).

Table 2
SIRM$_M$, S-Ratio and xARM/SIRM Means and Standard Deviation Measured on Bio-Sensor at Schools 1 and 2

School #1			SIRM (10^{-5} Am^2.kg^{-1})	S-ratio	xARM/SIRM (10^{-4} m.A^{-1})	N
Garland bio-sensors 13/12/2018 04/07/2019 I/O (no unit)		Indoor	1.35 ± 0.46	0.95 ± 0.01	9.14 ± 3.04	8
		Outdoor	1.88 ± 0.49	0.97 ± 0.02	4.83 ± 2.76	8
			0.7	–	–	–
Courtyard tree bark		West	6.40 ± 3.50	0.89 ± 0.07	–	7
		East	10.62 ± 3.26	0.92 ± 0.04	–	25
School #2						
Garland bio-sensors 20/12/2018–28/06/2019 I/O (no unit)		Indoor	0.95 ± 0.21	0.93 ± 0.05	5.69 ± 3.27	7
		Outdoor	1.08 ± 0.43	0.89 ± 0.05	5.31 ± 3.88	6
			0.9	–	–	–
Courtyard tree bark			8.61 ± 3.71	0.87 ± 0.05	–	18

Note. N is the number of samples and I/O is the ratio between indoor and outdoor SIRM$_M$.

almost stable throughout the experiments. In school 2, the value almost doubles from period 1 to 2. Outdoor EC concentrations are higher than indoor, and higher during period 1, reaching a maximum in School 1 (1.6 μg.m^{-3}). The minimum value is recorded for the outdoor environment in School 2 during period 2 (0.8 μg.m^{-3}).

The values of organic carbon (OC) concentration for the filters are higher indoors (Table 1), with a maximum value in School 1 of 5.5 μg.m^{-3} during period 2 and a minimum value in School 2 of 2.8 μg.m^{-3} in the same period. The outdoor values are maximum in School 1 during period 1 (4.4 μg.m^{-3}) and minimum in School 2 during period 2 (2.6 μg.m^{-3}).

The OC/EC ratio can be used to infer the source of the carbonaceous aerosols and also the formation of secondary organic carbon (SOC, Xu et al., 2015). The values observed in the schools are higher indoor than outdoor (Table 1). The indoor values have a small variation between the periods in School 1, with a maximum of 6.9 during period 2. In School 2, the OC/EC ratio decreases from 12.2 to 4.0 from periods 1 to 2. The outdoor ratio values have less variation, having a maximum in period 2 of 3.2 in both schools and a minimum at School 2 of 2.1 for period 1.

SIRM$_V$ measurements were carried out on the Teflon filters. It allows direct comparison between the concentration of iron oxides captured by the filters and PM1 concentration measured on the Quartz filters. SIRM$_V$ values in School 1 have maximum and minimum measured values of 3.21×10^{-10} and 0.49×10^{-10} A.m^{-1} in the outdoor environment in periods 1 and 2, respectively. In School 2, maximum and minimum values are reported outdoors during period 1 and indoors during period 2, with measured values of 1.98×10^{-10} and 0.42×10^{-10} A.m^{-1}, respectively.

3.2. Courtyard Tree Bark and Bio-Sensors

SIRM$_M$ values for the courtyard tree bark samples vary between 17.68×10^{-5} Am^2.kg^{-1} and 3.63×10^{-5} Am^2.kg^{-1} (Figure 1). Mean SIRM$_M$ values are close in all courtyards, ranging from $6.40(\pm 3.50) \times 10^{-5}$ Am^2.kg^{-1} (on School 1 West courtyard) to $10.62(\pm 3.26) \times 10^{-5}$ Am^2.kg^{-1} (on School 1 East courtyard). In School 2, tree bark SIRM values have a mean of $8.61(\pm 3.71) \times 10^{-5}$ Am^2.kg^{-1}. In School 1, a difference in the distribution of SIRM$_M$ values is noticeable between both courtyards, with higher values in the Eastern courtyard.

Bio-sensors were placed inside and outside the classrooms, on different floors of the building. SIRM$_M$ values for both indoor and outdoor bio-sensors of School 1 are higher than those obtained from School 2 (Table 2). In School 1, indoor bio-sensors have mean SIRM$_M$ values of $1.35(\pm 0.46) \times 10^{-5}$ Am^2.kg^{-1} - whereas mean values for outdoor samples are $1.88(\pm 0.49) \times 10^{-5}$ Am^2.kg^{-1}. SIRM$_M$ mean values for School 2 bio-sensors are $0.95(\pm 0.23) \times 10^{-5}$ Am^2.kg^{-1} (indoor) and $1.08(\pm 0.43) \times 10^{-5}$ Am^2.kg^{-1} (outdoor). The SIRM$_M$ mean values for bio-sensors display a similar trend than SIRM$_V$ and EC concentrations measured in the air filters, with higher values outdoors, in School 1.
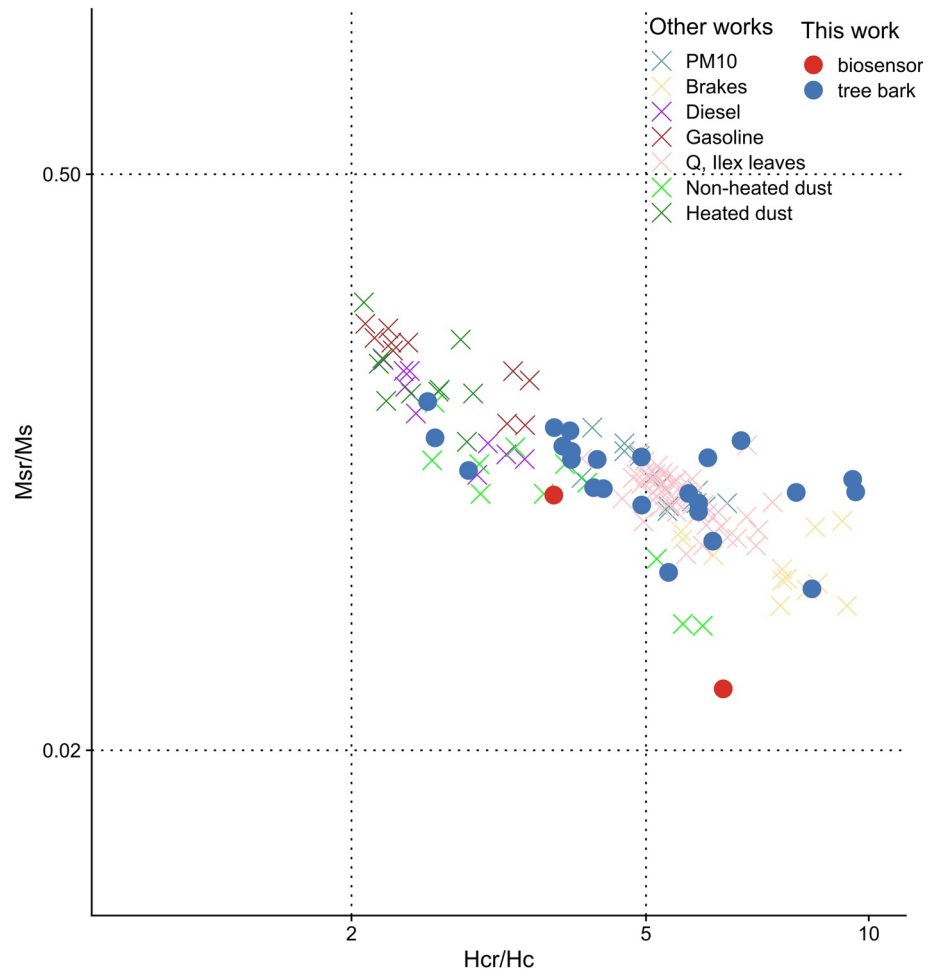
Figure 3. Day plot of the tree bark (circles) and previously published data (crosses). Data from Q. ilex leaves are from Szöyi et al. (2007), data for PM10 filters from Sagnotti et al. (2006), data for gasoline, brakes and diesel from Sagnotti et al. (2009) and Sagnotti and Winkler (2012), and data from dust (heated and non-heated) are from Górka-Kostrubiec et al. (2019).

The *S*-ratios calculated for the courtyards' barks display similar values for the three yards with 0.92($\pm$0.04), 0.89($\pm$0.07) and 0.87($\pm$0.05) for School 1 East and West ones and School 2 one, respectively. Results of the bio-sensors (Table 2) show a greater variability on School 2 when compared to School 1. Mean S-Ratio calculated for bio-sensors of School 1 are 0.95 ($\pm$0.01) and 0.97 ($\pm$0.02) in indoor and outdoor environments respectively. School 2 presents lower mean S-ratio values in comparison with the bio-sensors from School 1, with indoor and outdoor values at 0.93($\pm$0.05) and 0.89($\pm$0.05) respectively, indicating a higher content of high coercivity minerals than for School 1 samples.

Hysteresis parameters from both tree bark from the schoolyards' trunks and bark used as bio-sensors displayed a large range of parameters (Figure 3) compared, for example, to data obtained in Roma (Italy) on leaves (Szöyi et al., 2007) and on PM10 filters (Sagnotti et al., 2006). They extend to values obtained by Sagnotti et al. (2009) and Sagnotti and Winkler (2012) from exhaust residues from gasoline and diesel motors as well from brake wear dust collected directly in situ. This could result from different sources of particles, meaning different magnetic mineralogy such as high coercivity minerals like hematite(of geological origin e.g.,) or metallic iron (linked to abrasion of combustion cylinder, pads and disk brakes), which influences SIRM and coercivity values (Gorka-Kostrubiec & Szczepaniak, 2017; Gorka-Kostrubiec et al., 2019) and therefore impact the S-Ratio and/ or grain sizes parameters.

The xARM/SIRM values are found at the indoor environment of School 1 (Figure 4, Table 2), with a mean of 9.14($\pm$3.04) $\times$ 10^{-4}m.A^{-1}, whereas the outdoor ones have a mean of 4.83($\pm$2.76) $\times$ 10^{-4}m.A^{-1}.
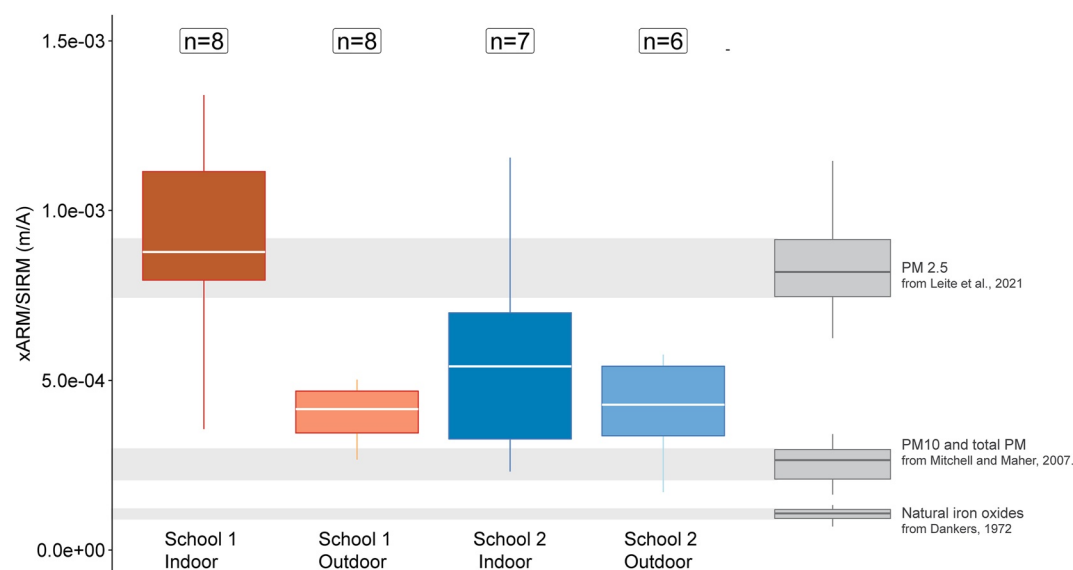
Figure 4. Box plot for calculated xARM/SIRM ratio in both school bio-sensors (indoor and outdoor) and bibliographical values (for natural iron oxides (Dankers, 1978); PM10 and total PM (Mitchell & Maher, 2009) and PM2.5 (Leite et al., 2021)). Data points in black dots, outliers in red dots, median is the horizontal line inside the box, the interquartile range by the size of the box, maximum and minimum values are the vertical whiskers.

School 2 displays similar calculated mean values in the indoor and outdoor environments, with values of $5.69(\pm3.27) \times 10^{-4}$m.A^{-1} and $5.31(\pm3.88) \times 10^{-4}$m.A^{-1}, respectively. School 2 mean values are also similar to the outdoor value of School 1.

The ratio of the indoor to the outdoor parameters (hereinafter called I/O; Table 2) is a simple and straightforward way to quantify the indoor-outdoor concentration relationship of a given compound, being used widely in air quality studies (Kalimeri et al., 2019 and references therein). I/O for EC shows similar behavior in both schools, increasing from period 1 to 2, with values of 0.4–0.8, and 0.3–0.9 for School 1 and 2, respectively (Table 2). Similarly, I/O for PM1 shows an increasing trend toward summer from 0.8 to 1.4 and 0.9 to 1.3, in School 1 and 2 respectively. I/O for SIRM$_V$ goes from 0.9 to 2.0 from period 1 to 2, showing more than a two-fold increase, whereas in School 2 there is a decrease from 0.5 to 0.2. The I/O for SIRM$_M$ on both school bio-sensors have close values.

3.3. SEM Observations

SEM results on the filters show the dense quartz matrix of the filters, encrusted with agglomerations of carbon "fluffy" soot aggregates, iron oxides aggregates and spherules. Figures 5a and 5b show backscattered electron images (BSE) from School 1 outdoor and indoor filters respectively (collected in period 2). For the outdoor filter (Figure 5a) one can notice an agglomeration containing iron (particle number 1), aluminum, and sulfur, with a dimension of 2.1 μm, and an elongated shape. Some carbon-rich agglomerates are also visible, for instance particle 2, 1 μm in size, mainly composed of carbon with some Na. Particle 3 is a 82 nm in diameter spherule, mainly composed of iron and oxygen. A secondary electron image (SE) with greater magnification (X40,000) (Figure 5a) enables further investigation of the topography of the iron oxide spherule (particle 3) and highlights the spherule and soot aggregate cementation to the quartz fiber.

The SEM image of the indoor filter (Figure 5b) shows an agglomeration of iron oxide spherules, attached within a carbon "fluffy" soot aggregate (dimension of 1 μm) on the quartz fiber. There are at least 6 spherules trapped (agglomerate 1), with sizes of 40, 57, 99, 123, 128 and 157 nm. Secondary electron image (Figure 5b) displays in greater detail the spherule agglomeration, cemented on the quartz filter fiber with soot aggregates.
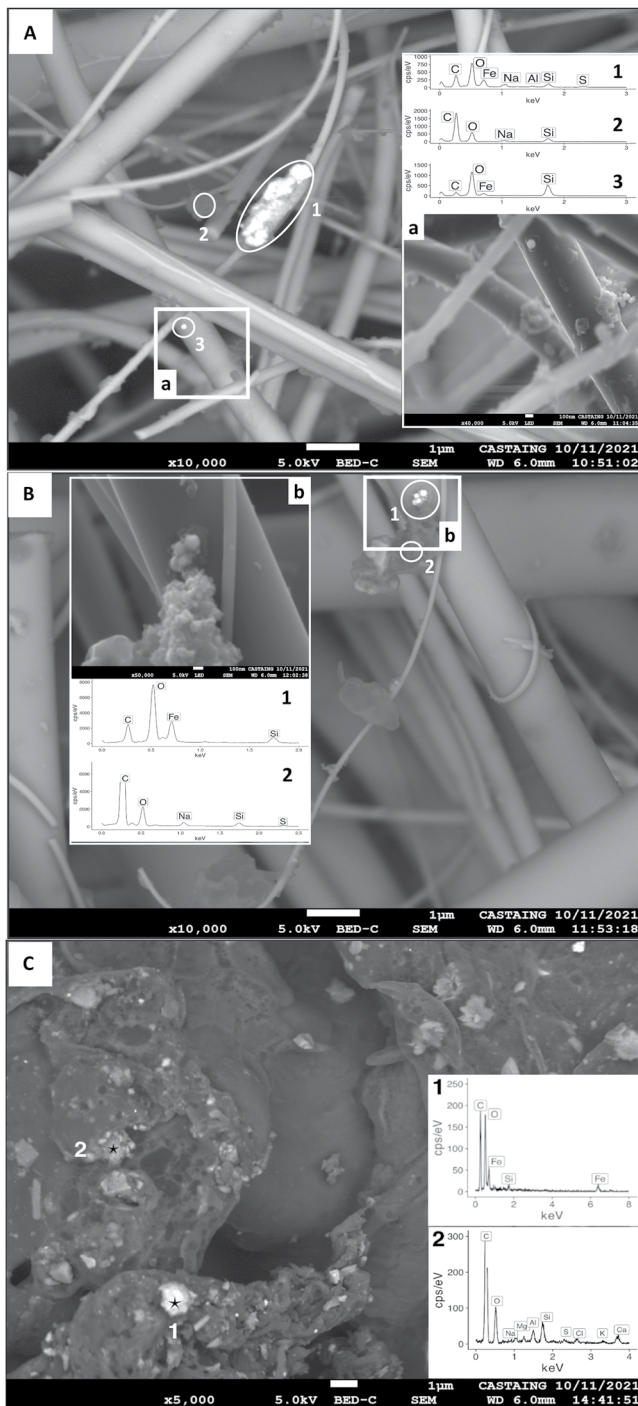
Figure 5. A: Backscattered SEM image of School 1 outdoor filter iron from period 2 with spectra of oxide spherule, iron oxide agglomerations and soot aggregates; (a) Magnified part of the image in Secondary electron image centered on the iron oxide spherule (particle 3). B: Backscattered SEM image of School 1 indoor filter from period 2 showing agglomeration of at least six iron oxides spherules (1) and carbon-rich aggregates (2); (b) Secondary electron image of agglomeration of iron oxide spherules (1); C: Backscatter SEM image from outdoor bio-sensors exposed in School 1 showing several agglomerated-like particles with irregular shapes including iron oxides.

Figure 5c displays typical particles from an outdoor bio-sensor in School 1. Particles are arranged in irregularly shaped agglomerates. Particle 1 is an iron oxide, with a dimension of 1.3 μm. Spectrum from particle 2 has a classical lithic composition (containing Mg, Al, Si, K and Na, Al and Si respectively) and size of 1.3 μm.

3.4. Teacher's Interviews

Two teachers were interviewed. One of the teachers reported that there are no official guidelines regarding air pollution except for avoiding outdoor sports during pollution peaks. Nevertheless, a poor air quality in the school environments linked to traffic emission is assumed by both teachers due to the visible proximity with the main traffic road for School 1 and an important access road to the city for School 2.

For the latter, occasional trash burning nearby the school is also mentioned. One of the teachers has a scientific background and says that his classrooms are receptive to topics concerning the environment. This teacher was involved in the design of the intervention by asking the researchers to come and explain the needs of children. From the teacher's point of view and observations, the fact that the researchers came to show the sensors made from bark, which is an inexpensive setup, was key in the appropriation of the project by the children. For this teacher, even if children didn't understand all the scientific background behind it, this is not problematic. What both teachers revealed is the importance that children were able to meet the researchers on more than one occasion, and to touch and to use the experimental material in their classrooms. Both teachers reported the problem of noise when the windows are open. Noises arise from children's activities in the schoolyard, helicopters and traffic, notably the honking of car and vehicle noises that reflect anger and stress. Besides the noise and difficulty of opening windows due to their dilapidated state, one of the teachers also reported some reluctance to vent in order to prevent traffic pollution from entering the classroom. The classroom is ventilated by the windows in the morning during cleaning before the arrival of children and sometimes during the day. Managing troubles in concentration and ventilating with an appropriate frequency is difficult for the teacher. The opening of windows during the day is induced by human sweat smelling and heating during the spring months.

Inconvenience due to the noise generated by the cyclone pump inside the classroom was reported by the pupils to their teachers after a few days of the experiment running. The teacher also reported relief at the end of the experiment with the active system despite the great motivation of pupils and teachers. Therefore, noise related to common active systems may prevent teachers' involvement in monitoring air quality in their classrooms while developing passive vegetal-oriented captors systems might be a way to develop their collaboration.

4. Discussion

4.1. Outdoor School Environment

The data indicates that the main source of the PM1 captured in both schools by the air filters is traffic emissions. Traffic emissions are generated by internal engine combustion and abrasion from the vehicle's parts (disk brakes for instance) and asphalt. Outdoors, Viana et al. (2007) reports OC/EC ratio values characteristic of traffic emissions in urban settings during winter and

summer for Belgium (4.4 and 3.5, respectively), The Netherlands (4.7 and 2.8, respectively) and Spain (5.6 and 4.6, respectively). In Athens, OC/EC ratios of 4.7 and 2.9 (Paraskevopoulou et al., 2014; Remoundaki et al., 2013, respectively) were found on traffic-related urban sites. Reported OC/EC ratios from outdoor samples in this study have a similar range compared with previous OC/EC ratios in European cities, pointing out traffic emission (specifically internal diesel combustion) as the main source of the captured PM1 in this study.

This traffic-sourced origin is further reinforced by the similarity of the measured trends obtained from the daily variations in NO_2 and PM2.5 concentrations (Figure 2). NO_2 is a secondary pollutant formed mainly from nitric oxide (NO), the predominant species in vehicle exhaust (Henderson et al., 2007). The covariation found between PM2.5 and NO_2 indicates that vehicle exhaust is the main source of PM2.5 in the city of Toulouse. The presence of iron-rich fly ash spherules among soot carbon chains (Figures 5a and 5b), on the PM1 filters, is also an indicator of internal combustion engine emissions found both indoors and outdoors (Leite et al., 2021; Shi et al., 2015). The presence of magnetite in PM is often linked to combustion and traffic sources, in line with the shapes detected for some iron oxides on the PM 1 filters (Figures 5a and 5b) by the SEM microscopy (spherules—Bardeli et al., 2011; Castañeda-Miranda et al., 2014; Gonet & Maher, 2019; Gonet et al., 2021; Leite et al., 2021; Liati et al., 2015; Sagnotti & Winkler, 2012; Winkler et al., 2021). The magnetic mineralogy of airborne iron oxides captured on tree bark and bio-sensors is composed of low coercivity carriers (magnetite-like) and high coercivity magnetic grains (e.g., hematite, goethite) as suggested by the S-ratio values between 0.89 and 0.97 (outdoors of Schools 2 and 1 respectively; Bloemendal et al., 1992; Frank & Nowaczyk, 2008). Some of the magnetic grains present in PM may have a geological origin, for instance being carried by wind over long distances (Formenti et al., 2014). Nevertheless, the magnetization used here (SIRM and ARM) are mostly dominated by the magnetite-like component.

Since traffic is the main source for PM in these school environments, the variability in SIRMs between courtyards, interpreted as variation of PM accumulation, could be caused by the impact of building configurations/architectural features (Figure 1). School 1 (West) and School 2 courtyards have similar configurations and similar SIRM values. Both present a tree line parallel to the main 3-floors school building making a corridor shape with the adjacent building that may increase venting. Conversely, School 1 (East) courtyard presents a u-shape configuration with the open side facing a sports ground and the ring road. The higher SIRM values observed in this courtyard could result from either the highest influence of the ring road (due to its proximity and open setting, Hofman & Samson, 2014; Hofman et al., 2013) and/or from the enclaved configuration limiting the venting.

Measured PM1 values in the schoolyards revealed a higher content in PM in both school environments than the background PM concentration from Toulouse monitored by official stations and especially during Period 1. This shows the influence from traffic emission sources in the studied schools due to their proximity to important major highways and access streets. Moreover, the two schools from Toulouse present comparatively high concentrations of PM1 compared to PM1 and PM2.5 data from European schools located in big urban centers (Figure 6). Outdoors, concentrations recorded in Toulouse are comparable to Western European cities such as Milan, Munich and Barcelona, all of which have larger populations (1.4 M, 1.5 M and 1.6 M respectively) than Toulouse (0.5 M). Each school has a particular context in a given city, explaining the large scatter in the recorded values (Figure 6).

4.2. School's Indoor Environments

As there are no indoor combustion sources, EC is a marker of outdoor air penetration. The I/O for EC increases between the first and the second period, indicating a higher influence of outdoor air in the classrooms with the changing of the seasons and temperature, due to more frequent opening of the windows as reported by the teachers. The mean I/O for EC for both sampled season periods equals 0.6 in both schools, similar to the one obtained from total magnetic PM measured on bio-sensors (0.6 and 0.9, respectively for School 1 and 2). The magnetic fraction of PM found indoors also points to external sources, hence traffic sources, in agreement with previous studies that found that magnetic particles on household dust are derived from traffic and industrial activities (Beata Górka-Kostrubiec & Iga Szczepaniak-Wnuk, 2017; Jelenska et al., 2017; Kelepertzis et al., 2019). Other known potential sources of magnetic PM indoors are cooking, combustion processes, smoking (Jordanova et al., 2006) and laser printing (ink composed of nanoparticles, Pirela et al., 2015), all of which are absent in the studied schools.
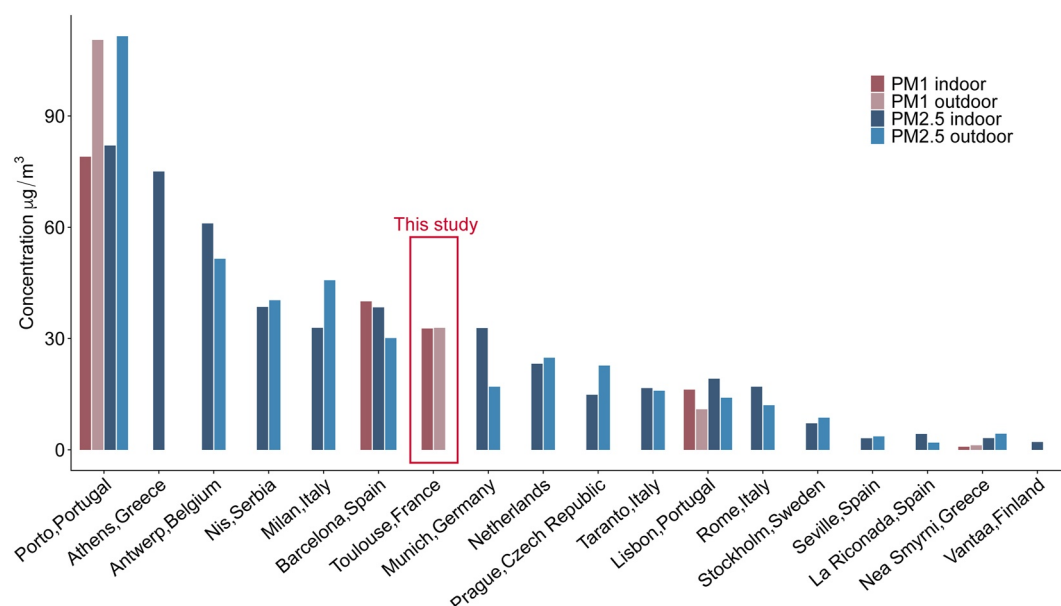
Figure 6. PM1 and PM2.5 mean concentrations in primary schools across Europe, in indoor and outdoor environments. See references from the supporting information.

High indoor values of OC concentration (Table 1), reflected on I/O for OC (Tables 1 and 2), point to indoor sources, such as organic emissions from children and staff, cotton fibers from clothes, wood furniture, crafting in school's activities (with the use of glue), among others (Branis and Sfranek, 2011; Fromme et al., 2008; Kalimeri et al., 2019; Rivas et al., 2014; Weschler & Shields, 1999). OC could also be generated indoor by reactions between oxidants from outdoors and gas-phase compounds emitted indoors, forming secondary organic aerosols (SOA) (Hodas et al., 2016: Long et al., 2000; Wainman et al., 2000; Waring, 2014; Waring & Siegel, 2010, 2013; Waring et al., 2011; Weschler, 2011; Weschler & Shields, 1999). Considering the high indoor OC concentration values reported in this study, especially during period 1 in School 2 and period 2 in School 1 (Table 1), the importance of proper ventilation on the school's environment becomes evident, given the potential toxicity of OC and SOC.

Proper ventilation is known to be an important issue in the school environment. In addition to the emission of particles, the accumulation of fine PM indoors (PM2.5, PM1) can occur in classrooms. Indeed, magnetic grain size distribution measured in School 1 displays a clear difference between indoor and outdoor environments, with a finer magnetic granulometry indoors close to the xARM/SIRM ratio of PM2.5 filters obtained by Leite et al. (2021). Conversely, School 2 samples present a similar magnetic grain size distribution indoor and outdoor typical of the entire PM range (Mitchell & Maher, 2009). School 1 outdoor environment also falls in this latter range. Although both schools have the same architecture, the teacher's interview revealed that all windows of School 2 were changed following the AZF industrial accident of 2001 allowing the windows to be fully open and classrooms properly ventilated, while School 1 has old tilt windows that can never be fully opened, which could have led to the differentiation in size range.

Indoor PM partly results from the penetration of PM from outdoor ambient air via openings (doors and windows) and building cracks (infiltration/exfiltration, Hodas et al., 2016). They can also be transported into the classroom by children and teachers (Chen & Zhao, 2011) via hair, clothes and bags. Here, high PM concentrations inside classrooms are related to poor venting. Indeed, the window ventilation is one of the most important actions to promote good air quality in schools (Almeida et al., 2011; Blondeau et al., 2005; Chithra & Nagendra, 2012, 2014; Rovelli et al., 2014) without automatic venting systems such as here, in Toulouse. The measurements of high CO_2 concentrations in School 1, required by law, also highlight a lack of ventilation especially in the instrumented classroom. A CO_2 concentration of 3,200 ppm is reported in 2018 for this specific classroom (Report of 12/06/2018) and over the 10 classrooms investigated, 7 were above 1,000 ppm. This is due to the aging of the openings and lack of renovation. In the classroom in concern, over the 10 existing windows, only five were still

functioning and only three could be effectively opened by the teacher. The teachers' interviews also indicated that the classroom is ventilated (opening the windows) in the morning during cleaning before the arrival of children and sometimes during the day. As also stated by Mumovic et al. (2009), managing troubles in concentration and the required ventilation is not easy for the teacher especially with absence of official guidelines regarding air pollution except for avoiding outdoor sports during pollution peaks. This study also highlighted that the teachers were not aware of the official CO_2 monitoring in their classrooms. The opening of windows during class time is described as subjecting children to noise, drafts and changes in temperature that have the effect of dispersing the group's attention.

High indoor concentration values of fine PM appear to be the hallmark of the school environment. Indeed, significant PM concentrations are recorded indoors especially in School 1 for period 1 as highlighted by high I/O for PM1 (Table 2) and SIRM values. High concentrations of PM2.5 have also been previously reported indoors in the school environment throughout Europe. A mean I/O ratio of 1.44 has been reported for 122 European schools (Kalimeri et al., 2019). Similar high values were obtained, among others, from 27 schools in Belgium with I/O ratios varying from 1 to 2.7 (Stranger et al., 2008) in 20 schools in Portugal with a mean I/O ratio of 1.45 (Madureira et al., 2012) or from 39 Spanish schools with I/O ratios varying from 1.2 to 1.5 (Rivas et al., 2015).

4.3. Vegetal Passive Collectors to Monitor Airborne Particles and Students-Teacher Involvement

The recorded concentrations in schools from Toulouse are higher than the air quality guidelines (WHO, 2021a) both outside and inside the classrooms. As one of the teachers explicitly stated, teachers and students suspected poor air quality because of the direct view and noise of the large entrance road next to the yard.

Both the informal exchanges between the teachers and researchers and the official interviews led to a co-design of the process and to the co-production of knowledge. Teachers were involved in the design of the intervention by asking the researchers to come more than once into the classrooms and explain the needs of children. From the teacher's point of view and observations, the fact that the sensor was made with almost nothing (verbatim *"trois fois rien"*) and from trees similar to those present in the yard was the predominant factor of endorsement by the children. The fact that children were able to meet the researchers several times, touch and experiment with concrete materials in their classrooms is the most important. Ecoanxiety as defined by Pihkala (2020) could affect all participants (Cunsolo et al., 2020; Dillon & Krasny, 2014; Russell & Oakley, 2016), the children as well as teachers and researchers. Both scientific activities around magnetic phenomena and the use of natural media help the discussion on air quality and its possible impact to be less anxiogenic for all actors.

For the monitoring of air quality in the school environment and the evaluation of children's exposure, the collaboration between the school (teachers, management and students) and the researchers makes it possible to better adapt the points of measurements via the feeling, acceptance and perceptions. Such an approach enables solutions or assessments to emerge. This empowers students and teachers to know their environment and to move toward solutions ranging from conscious acceptance to advocacy, legitimized by collaboration for practical measures. In this case, the co-building of solutions was initiated when preliminary results became available. Notably, they are concerned about the default in ventilation of the classrooms. However, the dialog was abruptly interrupted by the sanitary crisis of 2020.

5. Conclusions

A combination of magnetic and chemical methods highlights high ultrafine and fine PM concentrations of airborne particles both inside and outside the classrooms of two elementary schools in the city of Toulouse. Since the main source for PM is identified as traffic, both the proximity to the ring road and a lack of ventilation explain the measured values. Higher outdoor PM1 concentrations in both schools compared to the official traffic stations from downtown Toulouse point toward the fact that the local environment (i.e., distance to a source), here consisting of the proximity to the ring road and a major access road, is of crucial importance for the potential exposures of pupils. In addition, the microenvironments constrained by the architectural and vegetation configuration also have impacts on local PM concentrations. Collaboration between teachers, students, and scientists benefits the knowledge production about the influence of traffic sources in schools, school microenvironments, and environmental education. The use of vegetation as support for particles trapping, in addition to active sampling on filters, benefits both scientific studies by allowing the use of multiple sensors/stations and environmental education by

promoting school children's participation while reducing the anxiety associated with this type of environmental assessment. A comparison of PM accumulation in Toulouse and European schools shows a global tendency for higher PM concentrations in classrooms despite discrepancies in outdoor air quality. Epidemiological surveys inside school environments are urgently required to understand potential health and learning skills impacts on children of PM accumulation inside classrooms and would benefit from passive bio-sensors monitoring.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

Data set for this research is available in Zenodo repositories, under the https://zenodo.org/badge/DOI/10.5281/zenodo.6362617 (Leite et al., 2022).

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