



Research Paper

Plastic is in the air: Impact of micro-nanoplastics from airborne pollution on *Tillandsia usneoides* (L.) L. (Bromeliaceae) as a possible green sensor

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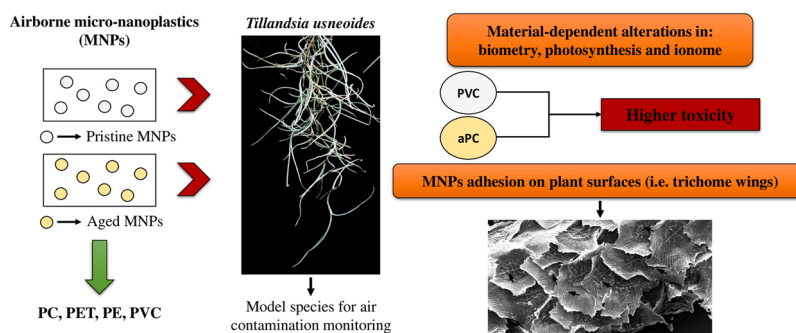
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HIGHLIGHTS

- Toxicity of airborne PE, PET, PC and PVC particles was tested on *Tillandsia usneoides*.
- Micro-nanoplastics were localised on plant surface, especially on trichome wings.
- Biometry, photosynthesis and ionome were altered in a material-dependent way.
- Pristine PVC and aged PC were the most toxic micro-nanoplastics for *T. usneoides*.

GRAPHICAL ABSTRACT



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ABSTRACT

Due to the increasing evidence of widespread plastic pollution in the air, the impact on plants of airborne particles of polycarbonate (PC), polyethyleneterephthalate (PET), polyethylene (PE), and polyvinylchloride (PVC) was tested by administering pristine and aged airborne micro-nanoplastics (MNPs) to *Tillandsia usneoides* for two weeks. Here we showed that exposure to pristine MNPs, significantly reduced plant growth with respect to controls. Particularly, PVC almost halved plant development at the end of the treatment, while the other plastics exerted negative effects on growth only at the beginning of the exposure, with final stages comparable to those of controls. Plants exposed to aged MNPs showed significantly decreased growth at early stages with PC, later in the growth with PE, and even later with PET. Aged PVC did not exert a toxic effect on plants. When present, the plastic-mediated reduction in plant growth was coupled with a decrease in photosynthetic activity and alterations in the plant concentration of macro- and micronutrients. The plastic particles were showed to adhere to the plant surface and, preferentially, on the trichome wings. Our results reported, for the first time, evidence of negative effects of airborne plastic pollution on plant health, thus raising concerns for related environmental risks.

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1. Introduction

Due to its convenient use, plastic is so widely employed that its debris are found all over our planet (Eerkes-Medrano et al., 2015; Dris et al., 2016; Bläsing and Amelung, 2018). The increasing presence of plastic in the environment, and its global distribution, make pollution deriving from polymeric materials among the most important environmental problems that our society must face, progressively reaching even nature sanctuaries and remote areas (Catarino et al., 2018; Nizzetto et al., 2016; Andrades et al., 2018; Scopetani et al., 2021). Research initially focused on very fine particles i.e. microplastics (0.1 μm to 5 mm) and nanoplastics ($< 0.1 \mu\text{m}$) involved in the contamination of water bodies and, successively, in soil, evidencing possible related risks to the various organisms that populate these environments (Zettler et al., 2013; Auta et al., 2017; Nava and Leoni, 2021; de Souza Machado et al., 2017; Wang et al., 2021; Scopetani et al., 2022). Currently, airborne micro-nanoplastics (MNPs) have growingly raised attention and concern (Dris et al., 2017), due to their increasing presence in the atmosphere, and their capability to undertake long-range transport. Several sources can contribute to the release of MNPs into the air (Catarino et al., 2018), thus generating a high heterogeneity in terms of chemical composition, and currently there are more than 5000 different types of plastics on the market (Zimmermann et al., 2019).

Fibres, foams, fragments and films are the most common shapes of plastic debris found in the atmosphere (Zhang et al., 2021). Their size has been found to range widely, from small particles around 20 μm to larger microfibers up to 5000 μm (Alfonso et al., 2021) and their abundance has been shown to reach fallout values of even 1000 items $\text{m}^{-2} \text{day}^{-1}$ (Wright et al., 2020). These particles are not only dangerous per se but can also represent vectors for other organic and inorganic contaminants (Wright and Kelly, 2017; Gasperi et al., 2018; Wright and Kelly, 2017; Rochman et al., 2014), with increased potential damage to living organisms (Gallo et al., 2018). In addition, the environmental persistence of polymeric materials not only makes plastic pollution an almost ever-lasting problem, but also exposes the particles to continuous aging processes, such as mechanical deterioration, photo-oxidation, chemical weathering and biological decomposition, thus altering their chemical structure, morphology, porosity and size (Hidalgo-Ruz et al., 2012; Kubowicz and Booth, 2017; Reynaud et al., 2022), and potentially increasing their toxicity (Luo et al., 2020).

Due to strengthened awareness that plastic debris is significantly dispersed in the air (Amato-Lourenço et al., 2020; Brahney et al., 2021), some studies have shown that these pollutants can reach animals and humans causing harmful effects (as reviewed in Enyoh et al., 2019 and Yang et al., 2021), including lung cancer (Prata, 2018). On the other hand, even though negative impacts of atmospheric microplastics are expected on the whole ecosystem, information about the potential threat of MNPs to organisms other than animals is extremely limited. In this context, land plants are fundamental primary producers and contribute to regulate the global climate (Duarte et al., 2013), therefore their response to airborne plastic particles should be better investigated. Indeed, plant leaves are one of the main targets of atmospheric pollution, since their large and uneven surfaces play a key role in intercepting a large portion of airborne particles, thus acting as a major sink (Bi et al., 2020). A recent statistical study estimated that leaf surfaces can retain about 0.13 trillion plastic pieces in the top 11 green countries (Liu et al., 2020). Therefore, insight into the impact of MNPs on plant leaves has implications for an accurate assessment of the threats of such materials to safety of our ecosystem. Actually, the negative impact of MNPs on plants have been reported from exposure via water and soil (Ge et al., 2021; Zhang et al., 2022). There are few reports showing that in the presence of MNPs aquatic and sediment-rooted macrophytes display depressed growth rate and reduced cell viability (Kalcíková et al., 2017; van Weert et al., 2019), changes in leaf functional traits, high ecotoxicity and oxidative damage induction (Yu et al., 2020). Regarding land plants, more information is available, with studies showing MNP-induced

reduction in shoot and root biomass, lowered germination rate, genotoxic and oxidative damage, depression of photosynthesis and changes in element composition and in metabolic profiles (see e.g. Boots et al., 2019; Jiang et al., 2019; Pignattelli et al., 2020; de Souza Machado et al., 2017; Colzi et al., 2022; Lian et al., 2020). Moreover, MNP treatment can affect the plant hormonal balance, with distinct effects in roots and shoots (Li et al., 2021; Zhou et al., 2021). Finally, while MPs negatively impact on plants mainly by altering the soil structure and microbiological characteristics, and through the direct contact with the root surface, NPs have been shown to enter the plant body owing to their sub-micrometre dimensions, thus exerting a direct toxicity once inside the plant tissues (Azeem et al., 2021).

In this work, the species *Tillandsia usneoides* (L.) L. was used to study the impact of airborne plastic pollution on plants. Among all organisms, plants belonging to the neotropical genus *Tillandsia* are valuable model systems for the estimation of contaminants present in the atmosphere, as they lack of a root apparatus and have a peculiar mechanism of water and nutrients uptake through leaf trichomes (Mosti et al., 2008). Trichomes are complex structures with an external wing composed of dead cells and a stem that transports water and nutrients to the trichome foot, which in turn finally distributes solutes to the underlying parenchyma (Papini et al., 2010). Since these plants absorb exclusively through the leaf trichomes and not through their roots, they have been used for biomonitoring atmospheric pollutants like metals and organic compounds (Brighigna et al., 2002). *T. usneoides* is the species of the genus that is easier to obtain since it lives in a wide range of environments, from Florida to Argentina (Sun et al., 2021), and is so common that it is used also as packaging material. For these reasons, this species was often used as a biomonitor of atmospheric pollution (Schreck et al., 2020).

The aim of this work was to test if MNP airborne pollution can negatively impact on plant growth. Accordingly, four different plastic materials were selected as model pollutants among those most frequently found in the atmosphere (Amato-Lourenço et al., 2020; Zhang et al., 2020), namely polyethylene (PE), polyvinylchloride (PVC), polyethylene terephthalate (PET) and polycarbonate (PC), and tested on *T. usneoides* plants cultivated in standard and controlled conditions. These materials also represent a broad spectrum of polymers from the point of view of synthetic procedures and chemical composition, which is expected to have an impact in the degradation pathway and aging mechanism. The MNPs obtained from the chosen plastics were administered to the plants in concentration and dimension ranges typically present in polluted air environments (Amato-Lourenço et al., 2020; Zhang et al., 2020). In addition, both pristine and aged plastic particles were tested, following the hypothesis that the aging process experienced by plastics during permanence in the environment might increase or decrease their toxicity, also depending on the starting material. This assumption is in line with recent reports from the literature (Guimarães et al., 2021; Wang et al., 2021; Latchere et al., 2021). The effects of these contaminants on *T. usneoides* were investigated through changes in plant growth, chlorophyll fluorescence and elemental profile. SEM microscopy was used to investigate possible MNPs adhesion to the plant surface.

To the best of our knowledge, this is the first report on the effect of airborne plastic pollution on plants, raising concern on a poorly known aspect of the diffusion of MNPs in the environment and claiming attention for its attenuation.

2. Materials and methods

2.1. Production and characterization of MNPs

The MNPs studied in this work were obtained by top-down procedures from plastic materials of common use: polycarbonate (PC, from a plastic sheet); polyethylene terephthalate (PET, from fruit packaging); polyethylene (PE, from a bottle); polyvinyl chloride (PVC, from chair leg caps). Our rationale was to include representative classes of polymers

with different chemical composition (Mark, 1999), in that the PC monomer is characterized by two aromatic rings bonded with a carbonate group that confers rigidity, the PET monomer contains an aromatic ring and two carboxyl groups, while PE and PVC are nonaromatic polymers, the latter of which also contain C-Cl bonds which are susceptible of homolytic cleavage and radical decomposition pathways (Brydson, 1999; Zaikov and Howell, 2006).

For all plastics, a sample of 0.5 g was firstly downsized with a sharp file to obtain a rough powder with particle diameters in the range 100–200 μm , and subsequently ground into a stone mortar. This allowed to obtain MNPs of 2–50 μm . The final size values are reported below (Section 3.2) and depended on the plastic properties, i.e. mainly on its hardness. To avoid overheating and partial fusion of the smallest particles, the grinding procedure was carried out in 5 steps, at the end of which the powders were homogenized with a stainless-steel spatula and an aliquot of each was analysed at the light microscope.

Self-contamination could potentially affect the evaluation of plastics content, in particular when synthetic fibers from personnel clothes (Scopetani et al., 2020) may accidentally pollute the samples at any stage of the process, i.e. collection, treatment and analysis of the plastics. In this work, all lab equipment (scissors, tweezers, etc), as well as personnel clothing and gloves containing plastics of the same type as those investigated, were avoided. Other possible sources of pollution were duly taken into account and subtracted by constant referencing to controls which were treated in exactly the same way as the samples. Furthermore, all glass and metallic surfaces of labware and instruments were cleaned with analytical grade acetone and dried with a N_2 flow prior to handling.

2.2. Weathering of plastic particles

To simulate the aging process that plastics undergo when released in the environment (Luo et al., 2020), 160 mg of the plastic powder were homogeneously placed in glass Petri dishes with 60 mm of diameter and submitted to accelerated aging for three weeks (preliminary trials indicated this exposure length as the first time span for significant changes to be detected in the MNPs with the analytical methodologies described below). The steps of this weathering procedure consisted of repeated cycles of 17 h exposure to UVC rays from a lamp with wavelength in the range of 250–280 nm and a net irradiance 2280 W m^{-2} , and a distance of 30 cm between the lamp and the samples (Lawal et al., 2017); 5 h exposure to light irradiance (lamp emitting at $\lambda = 290\text{--}800 \text{ nm}$) with light intensities ranging from 250 to 765 W m^{-2} after moistening (1.5 mL of tap water for each plate), and 2 h of heating at 60°C in an oven.

For the sake of simplicity, the aged particles (aMNPs) were named aPC, aPET, aPE and aPVC.

2.3. Pristine and aged MNPs characterization

Both pristine and aged MNPs were analysed using a Cary 620–670 Fourier transform infrared spectroscopy (FTIR) microscope (Agilent Technologies) equipped with an FPA (Focal Plane Array) 128×128 detector (Agilent Technologies), which allows performing 2D imaging-FTIR analysis; 128 scans were acquired for each spectrum, in reflectance mode with open aperture and a spatial resolution of 4 cm^{-1} . Background spectra were collected directly on a gold-plated surface. The Agilent Resolution Pro software (Agilent technologies) was used to collect, process, and analyse the spectra, and to obtain 2D imaging maps. Each analysis provides a “tile” map of $700 \mu\text{m} \times 700 \mu\text{m}$ (128×128 pixel) with an imaging map spatial resolution of $5.5 \mu\text{m}$ (i.e. each pixel has dimensions of $5.5 \mu\text{m} \times 5.5 \mu\text{m}$). The spectra showed in the FTIR images in Section “FTIR Reflectance spectra of pristine MNPs and aged MNPs” are representative of those obtained from tile maps collected on each MNP.

For the Scanning Electron Microscopy (SEM) analysis, the surface of

pristine and aged MNPs samples was covered with graphene using a sputter coater. Samples were analysed by a SEM Zeiss EVO 40 at MEMA (Centro di Servizi di Microscopia Elettronica e Microanalisi), the Service Center for Electron Microscopy and Microanalysis of the University of Florence. The instrument was set at 15 kV and at different working distances ranging from 6.0 to 5.5 mm.

To obtain the histograms of the MNP size distribution, images of the particles were taken with a Leitz DM-RB Fluo Optic microscope (Wetzlar, Germany) equipped with a digital camera Nikon DS-L1 (Tokyo, Japan). At 63x magnification. In particular, the size was measured by analysing six images for each plastic using Image J software. Each image had an area of $1.95 \times 10^{-2} \text{ mm}^2$, that was divided in six rectangular sectors of $3.25 \times 10^{-3} \text{ mm}^2$ and in each sector the size of five particles was measured (for a total of 180 measurements for each type of plastic material used).

2.4. Simulation of plant exposure to suspended atmospheric MNPs

To simulate MNPs spreading in the air, an ad hoc designed set-up was constructed as sketched in Fig. 1. A closed glass box (width: 30 cm, depth: 30 cm height: 20 cm) was used to expose nine *T. usneoides* strands of about 30 cm in length to airborne pollution (samples were folded up and hung to the lid by means of hooks). Air was moved by two fans (size $60 \times 60 \times 15 \text{ mm}$, direct current 12 V, speed 2500–6000RPM, 13 Cubic Feet per Minute CFM) directed towards the bottom of the box to create a turbulent motion. Before starting the experiment, the correct air circulation was checked observing the movements of little strips of soft paper after switching on the fans. The fans were switched on during the night, when the CAM species *T. usneoides* opens the stomata (Martin and Siedow, 1981), supposing a lower interaction of MNPs with the plant when such structures are closed. Nocturnal stomata opening was checked by a Leitz D.M.-R.B. light and fluorescence microscope equipped with a Nikon DS-5 M digital camera. The fans kept working for three hours, since a preliminary screening indicated that it was the maximum time of ventilation allowed before inducing plant desiccation. Two beakers filled with demineralized water were placed in the box to maintain humidity, and two openings with circular filters (diameter of 4.5 cm, porosity $0.3 \mu\text{m}$) were fixed at the two opposite sides of the box to allow air flux in the box without loss of MNPs. Finally, these box prototypes were placed in a growth chamber ($24/16^\circ\text{C}$ day/night; light intensity $200 \mu\text{mol m}^{-2} \text{ s}^{-1}$, 16 h (day) photoperiod; relative humidity 60–65%).

Tillandsia usneoides plants were purchased at a local plant nursery (Giulio Celandroni Orchidee, San Giuliano Terme, Pisa) and maintained in a greenhouse of the Botanical Garden of Florence for 2 months prior to the experiments to guarantee that all specimens were healthy and pest-free. At the beginning of the experiment, strands were obtained from mother plants and some of them were SEM analyzed as described below to make sure that no exogenous particles were adhering to the plant surface. Strand length was measured and samples of homogeneous size and with similar number of ramifications (around 5) were chosen for the trial. For length measuring, images of the plants were analyzed by ImageJ (<https://imagej.nih.gov/>), all the ramifications were measured, and a cumulative length of the plant was calculated. The stem diameter was constant.

Tillandsia usneoides strands were exposed to levels of MNPs reported in highly contaminated outdoor places i.e. in the order of $10^4 \text{ items} \cdot \text{m}^{-3}$ (Zhang et al., 2020; Asrin and Dipareza, 2019). The calculation for assessing the MNP amount to be allotted in each prototype chamber was based on an average density of the four polymers used in this work (PE = 0.94 g cm^{-3} ; PET = 1.38 g cm^{-3} ; PC = 1.2 g cm^{-3} ; PVC = 1.4 g cm^{-3}) and the volume of the particles. These were assumed to have spherical shape of known mean diameter, whose value was extracted as the mean of the corresponding size distribution. The heavily polluted atmosphere was replicated in a box of 18 dm^3 by administering a total of 7, 10, 9, 10 μg for PE, PET, PC and PVC respectively (weighed by a Sartorius

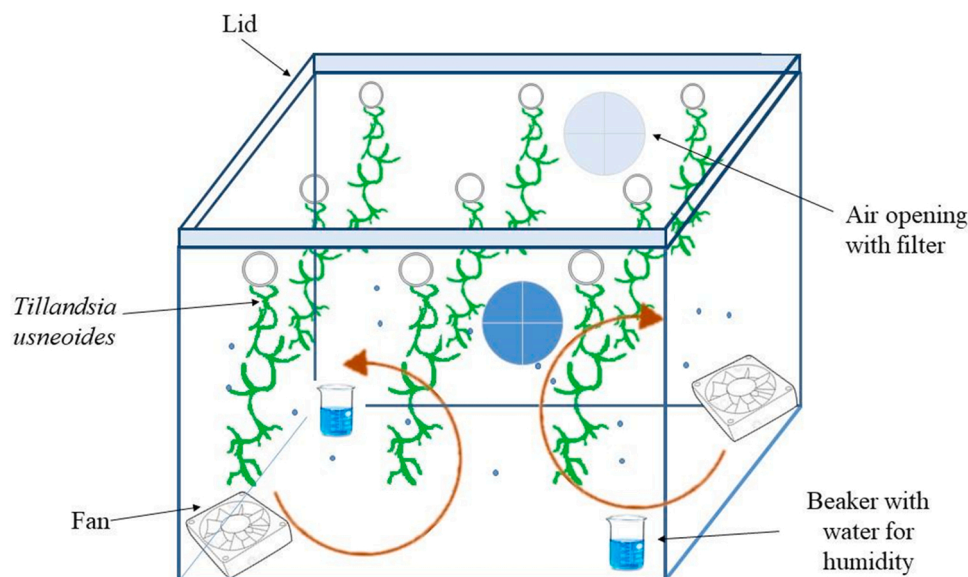


Fig. 1. Scheme of the set-up used in the air circulation experiments.

Cubis® II Ultra-Micro Lab Balance). MNPs powders were placed on a glass slide under fans before switching the fans on. Thus, the fans with downwards direction promoted a circular movement of the MNPs into the box. During MNPs exposure, each *T. usneoides* strand was hydrated with 5 mL of sprayed tap water three times a week.

During the exposure, increment in plant growth was calculated by subtracting length values at the beginning of the treatment to values at 4, 7, 11, 14 days of treatment. The experiment was stopped after 14 days of exposure, since this was the first point in time when the pattern of differences in plant growth caused by the MNPs did not change from the previous measurement.

2.5. Fluorescence parameters

Chlorophyll a fluorescence was measured on 15-min dark-adapted leaf samples using a portable fluorimeter (Plant Efficiency Analyzer – Handy PEA, Hansatech Instruments Ltd). Samples were flashed for 1 s with a saturated ($1800 \mu\text{mol m}^{-2} \text{s}^{-1}$) red light pulse (650 nm) from a LED into the fluorimeter sensor. Leaf sample health was evaluated using the potential quantum efficiency of photosystem II, i.e. the indicator of photosynthetic efficiency F_v/F_m (F_v indicates the difference between the maximal F_m and the minimal F_0 fluorescence), and by the performance index (P_{index}), which are the general indicators of photosystem I and II efficiency.

2.6. Element determination

At the end of the experiment, plants were carefully washed with demineralized water and dried at 70°C for two days. The concentration of micro- and macro-elements was measured by wet-digestion of 100 mg of oven-dried plant material using 10 mL of 69% HNO_3 in a microwave digestion system (Mars 6, CEM) with maximum temperature of 200°C for 10 min (Bettarini et al., 2019). Atomic absorption spectroscopy (PinaAcle 500, Perkin Elmer) was used to determine the amount of K, Ca, Mg, Fe, Zn, Cu and Mn, and concentrations were expressed on a dry weight basis. Certified reference materials (grade BCR, Fluka Analytical, Sigma-Aldrich) were used to verify the methods reliability and accuracy (values $< 10\%$ and $< 5\%$ RDS respectively).

2.7. Microscopy

Scanning Electron Microscopy (SEM) analysis was conducted as in

Papini et al. (2010): fragments of leaves were fixed in a solution of formaldehyde 10%, Ethanol 50%, Acetic Acid 5% (FAA) and then, after a washing in phosphate buffer (pH 6.8) 0.1 M, dehydrated in a series of increasing concentrations of ethanol (70%, 80%, 95% and 100%). After dehydration, the samples were critical point dried and gold metalized using a sputter coater. Finally, samples were observed with a Zeiss EVO 40 SEM by the MEMA center of the University of Florence (Centro di Servizi di Microscopia Elettronica e Microanalisi – MEMA). The instrument was set at 15 kV and at different working distances ranging from 6.0 to 5.5 mm.

To assess the distribution of MNPs on the plant surface, a count of the particles present on the shield and on the wing of the trichomes was performed. For each SEM images, three trichomes with the whole surface visible horizontally were selected and particles were counted on an area of $10000 \mu\text{m}^2$ randomly located on the shield and on the wing. Values were pooled together to obtain a mean value for each image. For each plant, three SEM images were analysed, and their mean was considered as the final value of the replicate.

2.8. Statistics and PCA

The experiment comprised nine groups of plants (control plants, plants treated with the four kinds of MNPs, plants treated with the four kinds of aMNPs) and for each group there were nine biological replicates. When measurements were performed over time (plant length and fluorescence parameters), the same plant was evaluated at different times.

Experimental data of plant growth were analysed by linear regression (using GraphPad Prism 7, GraphPad Software, San Diego, CA), where plant length increment was the dependent variable and sampling time the independent time. The angular coefficient (slope) of the linear regression was assumed as a descriptor of the plant growth rate.

One-way ANOVA was used to check the significance of differences ($p < 0.05$) among means at the same experimental times (and among the slope values), using GraphPad Prism 7 (GraphPad Software, San Diego, CA). A HSD-Tukey test was run for post-hoc comparisons. Data normality was checked with the Shapiro-Wilk test.

The data of plant length increment, F_v/F_m and P_{index} were analysed fitting linear mixed-effects models (LMMs) in a repeated measurement ANOVA design, using plant identity as a random effect factor to account for the temporal correlation of observations. Time was used as an ordinal variable. The physiological effects of the treatment were

investigated using plant length increment, F_v/F_m and P_{index} values as response variables and the treatments with plastic particles as explanatory variables in a full factorial design. ANOVA type III table, with Satterthwaite's method was used to verify the significance of the fixed effects and of associated interaction factors. The analyses were run with the free software R. LMM computations were performed using the lmer function of the lme4 package version 1.1–12 for fitting the models.

The relationships among MNP treatments and plant traits (increment in length, F_v/F_m , P_{index} and element concentration) in *T. usneoides* plants were evaluated using Multiple Factor Analysis (MFA) implemented in the package FactoMinerR (Lê et al., 2008) under the R software v.4.1.3 (R Core Team, 2020). The Principal Component Analysis (PCA) was performed to summarize the variation of *T. usneoides* plants treated with pristine and aged MNPs. Two separate MFA were performed considering the ageing of the used plastics.

3. Results

3.1. Pristine and aged MNPs characterization

SEM micrographs of pristine MNPs showed that the largest fraction of items was in the range of 1–10 μm ; two small fractions were over 20 μm and less than 1 μm , respectively (Fig. 2A).

The aPC particles showed more uniform size distribution compared to the pristine MNPs with dimensions in the range of 0.5–2 μm , and only a small part having more than 30 μm size (Fig. 2B).

Pristine PET particles showed larger size (30–50 μm) than aPETs, which were smaller than 10 μm (Fig. 2C) with a large portion of sub-micron dimensions (less than 0.5 μm , Fig. 2D).

Regarding PE, the MNPs size was mainly around 2–3 μm (Fig. 2E), with a small portion having dimensions 30–50 μm , and aPE particles

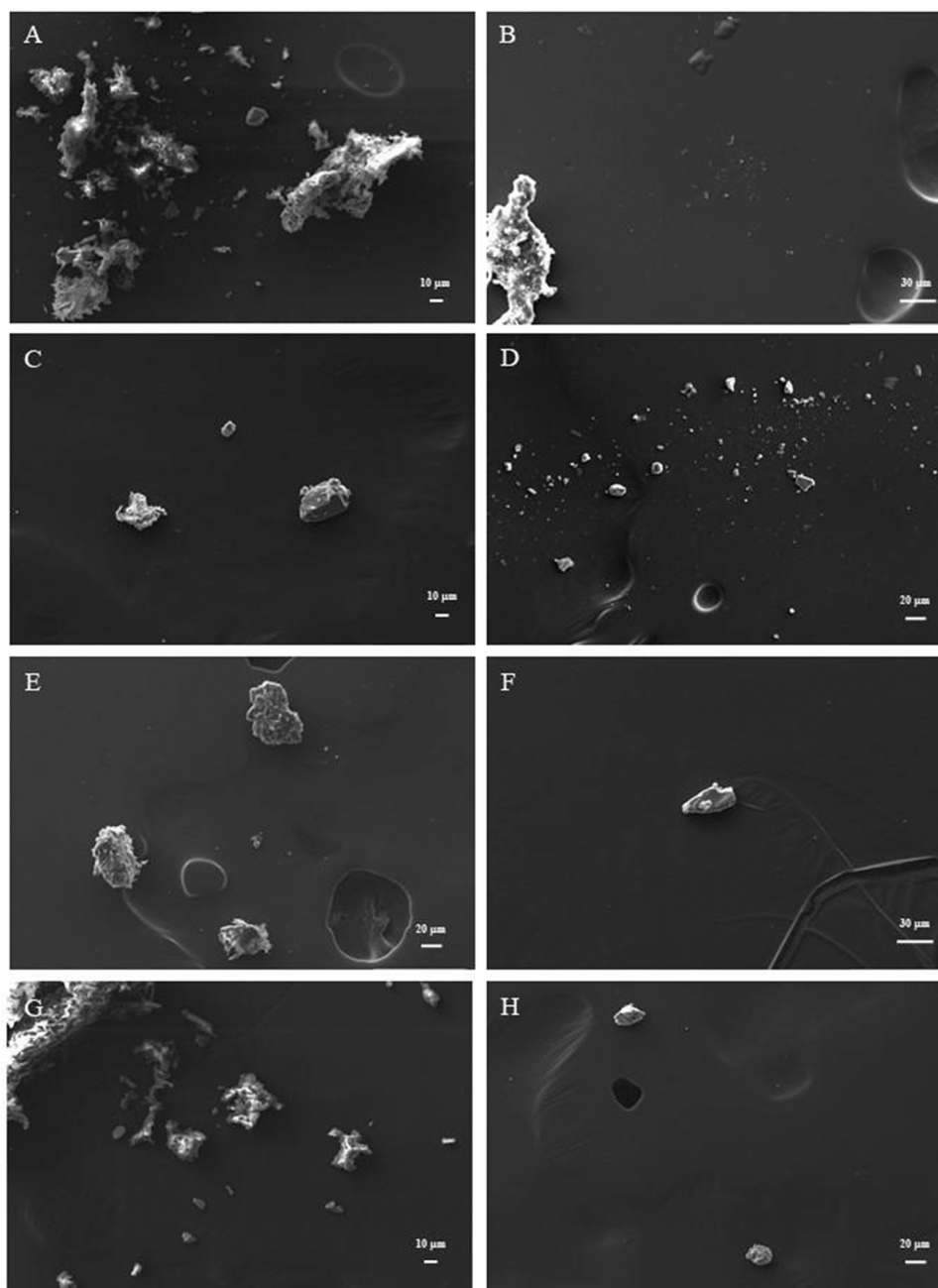


Fig. 2. Representative SEM micrographs of (A) pristine PCs (Mag 693x), (B) aPCs (Mag 693x), (C) pristine PETs (Mag 438x), (D) aPETs (Mag 548x), (E) pristine PEs (Mag 438x), (F) aPEs (Mag 558x), (G) pristine PVCs (Mag 693x), (H) aPVCs (Mag 438x). Mag = Magnification.

showed comparable sizes to their pristine counterparts (Fig. 2F).

PVCs showed variable sizes spanning from 2 μm to 25 μm (Fig. 2G), similar to the aged particles (Fig. 2H).

The histograms of the size of pristine and aged MNPs are shown in Figure S2. These size distributions were all well reproduced by a log-normal monomodal function (also reported in Figure S3), in agreement with previous studies performed on environment polluting microplastics (Kooi and Koelmans, 2019). The corresponding mean, standard deviation, coefficient of variation median are reported in Table S1. From the size histograms it appeared that the curves of

weathered particles are slightly shifted toward higher values with respect to the corresponding pristine specimens. This size increase could be due to the Ostwald ripening, a thermodynamically driven process commonly found in dispersed systems, which occurs upon aging to reduce the interfacial free energy (Ruth et al., 2003; Xu, 2014).

3.2. FTIR reflectance spectra of pristine MNPs and aged MNPs

The FTIR analysis of the MNPs evidenced changes in the chemical composition and structure of the polymers submitted to accelerated

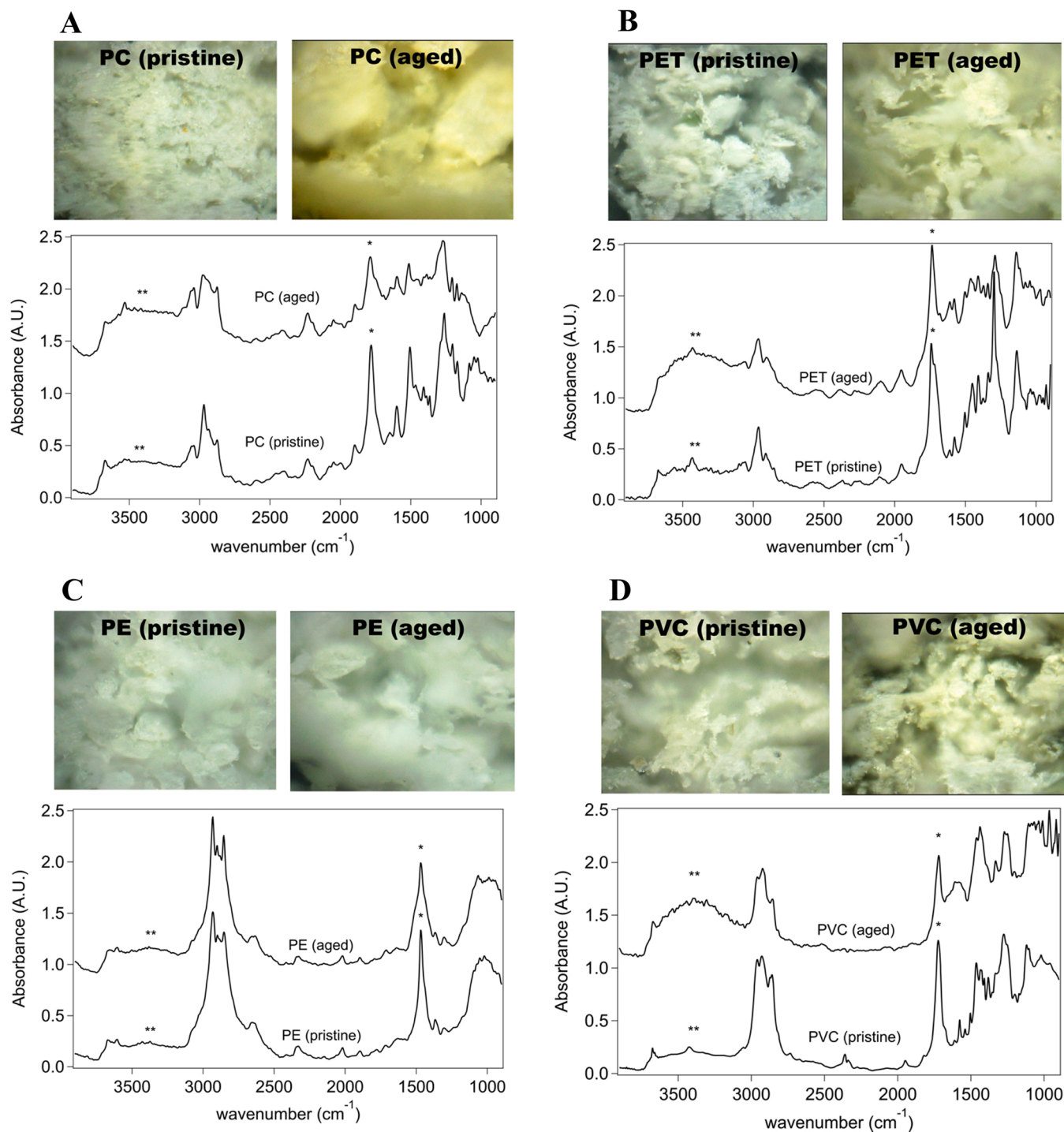


Fig. 3. Visible light microscopy images and FTIR Reflectance spectra of PC (A), PET (B), PE (C), and PVC (D) samples before (pristine) and after accelerated aging (aged). The stars highlight the OH stretching (****) and the C=O stretching (**) spectral regions.

aging (Fig. 3A-D). A marked intensity increase in the OH stretching region was observable in all cases, and it was particularly evident for aPET, aPE and aPVC. This finding was ascribed to oxidation upon exposure to UV light with contribution from humidity retained by the polymers. The yellowing observed in the visible images of aPC and aPET (and to a lesser extent also aPVC) is consistent with the formation of conjugated double bonds formed by the thermal transformation of oxidized groups produced during aging. Significant band broadening was also observed in the carbonyl (C=O) stretching band of aPC and in the CH₂ bending band of aPE, and it was explained with structural rearrangements and disorder increase in the polymers. Details of the OH stretching intensity increase in aPET, and CH₂ band broadening in aPE, are provided in Figure S1, where changes in IR absorbance of the two bands are mapped on the surface of the polymer powders/beads with spatial resolution of 5.5 μm (pixel size = 5.5 $\times$ 5.5 μm^2) and a detection limit in the order of less than 1 pg/pixel (Mastrangelo et al., 2020) thanks to the FPA detector of the FTIR microscope. In the case of aPVC, decrease in the intensity of the carbonyl bands of plasticizers (e.g. phthalates, citrates or adipates) was ascribed to their partial loss and release during aging. Similarly, the decreased intensity of absorptions in the 1600–1550 and 1250–1200 cm^{-1} regions in aPC might indicate the partial loss of Bisphenol A, which can be released from PC even at room temperature (Howdeshell et al., 2003).

3.3. Impact of MNPs on plant growth and photosynthetic parameters

Regarding the increment in length (Figure S4), at each time of the experiment one-way ANOVA highlighted significant differences among the treatments ($25.34 < F < 84.23$ and $P < 0.0001$ for all the times of treatment). In particular, control plants showed an increasing increment in length over time, whereas exposure to pristine MNPs caused a significant reduction in plant growth with respect to control plants after four days in samples treated with all the studied materials, except PVC. After seven days the plant growth reduction was significantly evident for plants treated with PET and PVC, and, at the other times monitored during the experiment, with PVC only (Figs. S4, 4 A). Regarding aged MNPs, a significant reduction in the increment of plant length with respect to the control plants was present in samples treated with aPC starting from four days of treatment (Figure S4). By increasing the duration of exposure, such reduction became significant also for aPE starting from seven days, and for aPET after 11 days. At the end of the treatment, the reduction in growth was significant for all the treatments

except aPVC (Fig. 4 A). Length increment variation changed significantly depending on the plastic material used for the treatment (treatment/time significant with $P < 0.001$, Table S2). As the regression analysis gave significant results for the correlation between increment in length and sampling time (Fig. 4B, S4), the angular coefficient (slope) of this regression line was used to estimate the growth rate itself. This coefficient showed significant differences among the treatments ($F=329.3$, $P < 0.0001$). PE, PET, PC and aPVC-treated plants showed slope values similar to that of control plants, while the other treatments displayed lower values, with aPC-treated samples showing the lowest.

As for chlorophyll fluorescence, one-way ANOVA highlighted significant differences for the treatments at each time of the experiment ($3.85 < F < 65.19$ and $P < 0.001$ for F_v/F_m and $9.79 < F < 26.86$ and $P < 0.0001$ for P_{index} for all the times of treatment). Specifically, in samples exposed to the pristine plastic materials, a significant decrease in F_v/F_m and in P_{index} with respect to control plants was induced only by PVC at all the treatment times starting from four days of exposure (Figs. 5 and 6). In the case of aMNPs, after four and seven days of treatment, the values of F_v/F_m were significantly lower in aPVC and aPC treated plants with respect to the controls, showing lower values also of P_{index} in the case of treatment with aPVC at four days and with aPC at seven days (Figs. 5 and 6). When evaluating the effects at longer exposure times, at 11 days of treatment all the aged plastic materials, except aPVC, exerted a significant negative effect on both the photosynthetic parameters, whereas at the end of the exposure aPC was the only pollutants causing a significant decrease in the values of F_v/F_m and P_{index} . The variation over time of the two photosynthetic indexes significantly depended on the kind of MNP used for the treatment (treatment/time significant with $P < 0.001$ for both: Table S3 and S4).

3.4. Microplastics alterations on the concentration of macro- and microelements

The effects of the four pristine and aged MNPs on the concentration of some macro- and micronutrients elements are reported in Table 1. One-way ANOVA highlighted significant differences among the treatment for all the element tested ($7.154 < F < 146.6$ and $P < 0.001$ for all the metals). In particular, microplastic exposure did not significantly alter the levels of the elements in case of MNPs, except when PVC was used for treatment. This material induced a general decrease in the concentration of all the studied elements, that was significant for Potassium (K) and Magnesium (Mg) among the macronutrients, and for Iron (Fe), Zinc (Zn) and Copper (Cu) among the micronutrients. Concerning aMNPs, strands of aPE- and aPC-treated plants showed a significant increase in the level of all the macronutrients, aPET-treated plants only in the case of Mg, whose level was instead decreased by treatment with aPVC. As for micronutrients, aPVC exposure significantly decreased the plant concentrations of all the studied elements. aPE and aPC treatment induced an increase in Zn concentration and a decrease in Manganese (Mn) in *T. usneoides* plants, whereas aPET exposure provoked a rise in the Mn level.

The amplitude of the treatment-induced changes in element concentration was plastic-, age- and element-dependent, as represented by the heatmaps (Fig. 7), where red colour denotes samples with decreased element concentration in respect to the corresponding control and green colour denotes samples with increased element concentration in respect to the corresponding control. In general, the variations were less evident in case of MNPs, except for PVC, whereas they were found to be remarkably manifest in aMNP-treated samples, with aPET showing the mildest impact. Particularly, the strongest effect among all the treatments was induced by PVC and aPVC, which caused the greatest decrease of K and Cu, respectively, in the strands, and by aPC, which induced the widest increase in Zn concentration.

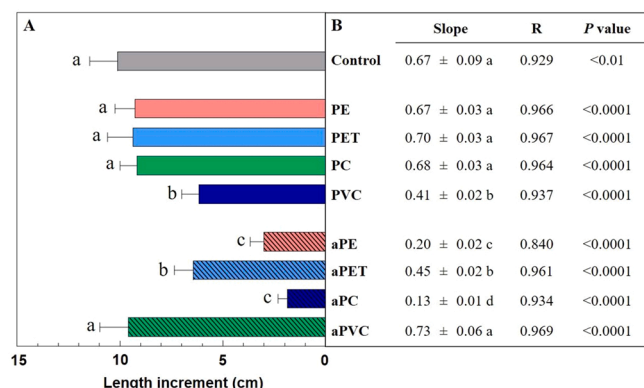


Fig. 4. A) Final increment in length (cm) of *Tillandsia usneoides* strands exposed to four types of pristine and aged MNPs for 14 days. Increment was calculated subtracting length values at the beginning of the treatment to values at the end of the treatment. Letters close to the histograms indicate the significant differences among the treatments according to the Tukey's test (at least $p < 0.05$). Values are means of 9 replicates $\pm$ standard deviation. B) Growth rate calculated as the slope of the linear regression between plant length increment and sampling time.

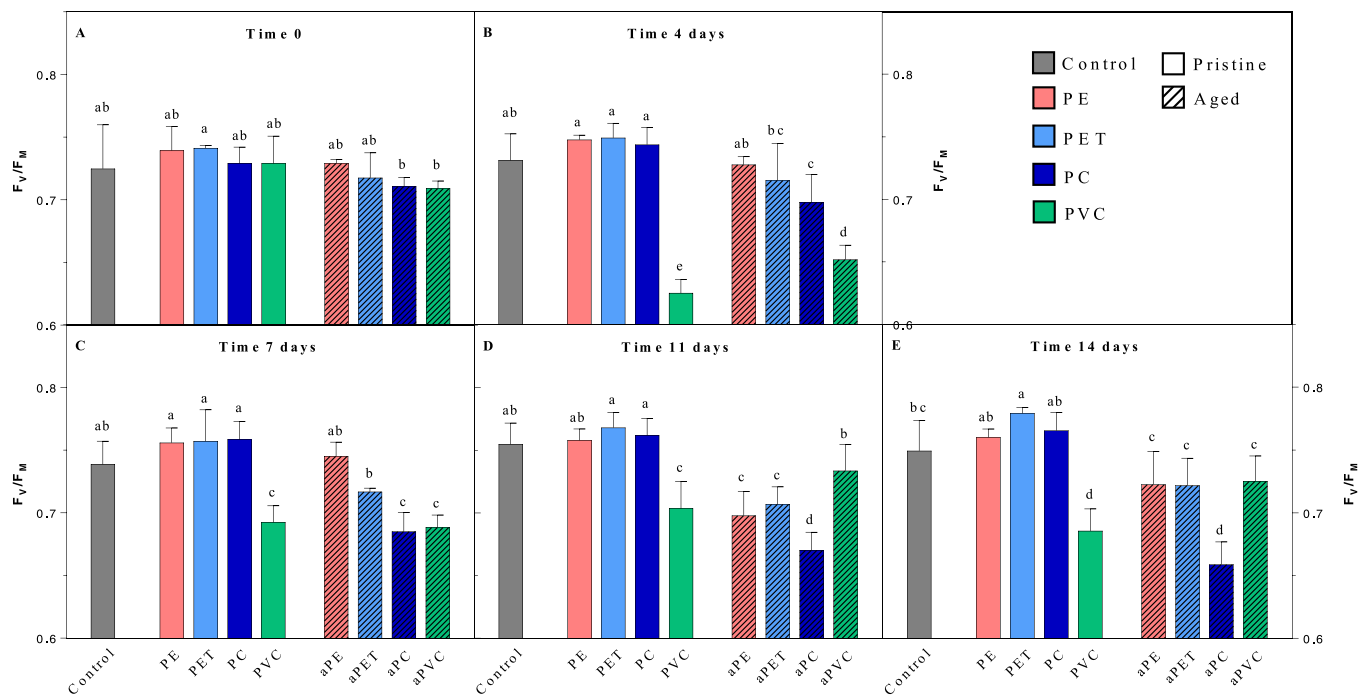


Fig. 5. Values of Variable Fluorescence/Maximal Fluorescence (F_v/F_m) of *Tillandsia usneoides* strands exposed to four types of pristine and aged MNPs at A) 0 days, B) 4 days, C) 7 days, D) 11 days and E) 14 days after the beginning of the treatments. Letters above the histograms indicate the significant differences among the treatments according to the Tukey's test for each time of treatment (at least $p < 0.05$). Values are means of 9 replicates + standard deviation.

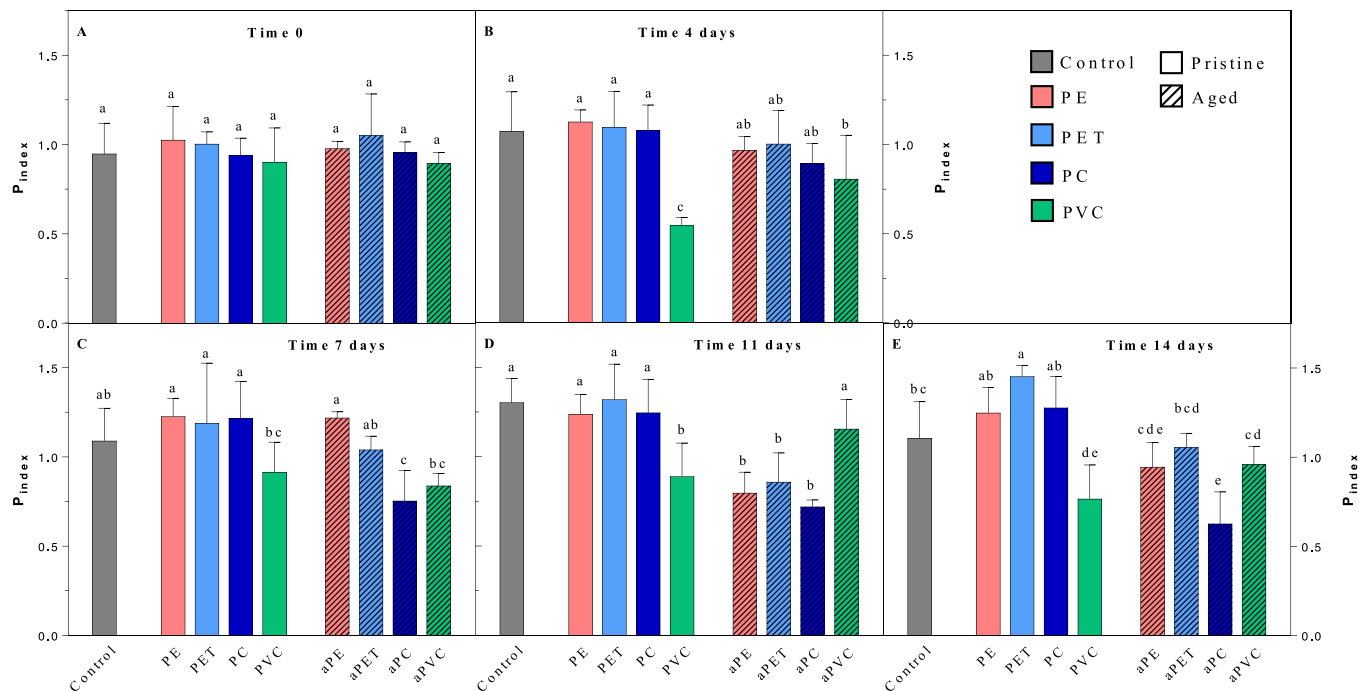


Fig. 6. Values of Performance Index (P_{index}) of *Tillandsia usneoides* strands exposed to four types of pristine and aged MNPs at A) 0 days, B) 4 days, C) 7 days, D) 11 days and E) 14 days after the beginning of the treatments. Letters above the histograms indicate the significant differences among the treatments according to the Tukey's test for each time of treatment (at least $p < 0.05$). Values are means of 9 replicates + standard deviation.

3.5. PCA results

PCA was performed for 10 main traits (plant length increment, 2 photosynthetic parameters, concentration of 7 metal ions in plant) in specimens treated with pristine and aged plastic particles. Considering all variability, PC1 retained 52.2% and 60.9% of the data variation for

MNP and aMNP-exposed samples, respectively, while PC2 retained 10.5% and 13.5%, as reported in the scattergram for pristine (Fig. 8A) and aged (Fig. 8C) MNP-treated plants. The direction and length of the vectors were substantially different, with the scattergram of the aMNP-treated plants showing longer vectors spread over both the first and the second sector with respect to the MNP-treated plants. Pristine and aged

Table 1

Element concentration ($\mu\text{g g}^{-1}$ dry weight) in *Tillandsia usneoides* plants after 14 days of growth in presence of pristine and aged MNPs. Values are mean of 9 replicates $\pm$ standard deviation. Asterisks indicate significant differences between treated samples and control samples (* = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$).

Treatment	K	Ca	Mg	Fe	Zn	Cu	Mn
Control	11,451 $\pm$ 2682 b	20,594 $\pm$ 1516 b	5552 $\pm$ 311 d	113 $\pm$ 21 a	161 $\pm$ 24 c	6.3 $\pm$ 0.8 a	38 $\pm$ 4 b
PE	11,173 $\pm$ 1531 b	19,105 $\pm$ 1667 b	5399 $\pm$ 462 de	114 $\pm$ 17 a	159 $\pm$ 29 c	6.1 $\pm$ 0.9 ab	41 $\pm$ 6 ab
PET	10,959 $\pm$ 2247 b	20,739 $\pm$ 1434 b	5608 $\pm$ 373 cd	112 $\pm$ 19 ab	168 $\pm$ 26 c	6.7 $\pm$ 0.8 a	39 $\pm$ 5 ab
PC	11,256 $\pm$ 1624 b	21,006 $\pm$ 1533 b	5491 $\pm$ 457 de	117 $\pm$ 14 a	154 $\pm$ 21 c	6.6 $\pm$ 0.7 a	37 $\pm$ 7 bc
PVC	7152 $\pm$ 1206 c	18,910 $\pm$ 1428 b	4886 $\pm$ 140 e	79 $\pm$ 2 c	112 $\pm$ 3 d	5.0 $\pm$ 1.2 b	36 $\pm$ 5 bc
aPE	14,114 $\pm$ 1901 a	25,503 $\pm$ 1690 a	7323 $\pm$ 1244 ab	131 $\pm$ 13 a	233 $\pm$ 27 b	6.2 $\pm$ 1.2 ab	27 $\pm$ 3 d
aPET	12,201 $\pm$ 721 ab	21,785 $\pm$ 1891 ab	6131 $\pm$ 512 bc	107 $\pm$ 16 ab	169 $\pm$ 6 c	4.9 $\pm$ 2.1 ab	44 $\pm$ 6 a
aPC	15,339 $\pm$ 620 a	27,548 $\pm$ 5937 a	7784 $\pm$ 1766 a	138 $\pm$ 35 a	374 $\pm$ 4 a	6.2 $\pm$ 0.4 ab	27 $\pm$ 2 d
aPVC	10,085 $\pm$ 4470 bc	18,829 $\pm$ 3025 b	5038 $\pm$ 475 cd	86 $\pm$ 5 bc	118 $\pm$ 13 d	3.7 $\pm$ 1.1 b	32 $\pm$ 5 cd

MNP treatments for *T. usneoides* in the PCA of individuals are showed in Figs. 8B and 8D, respectively. The main feature in the distribution of the MNP-treated samples was that the group of PVC-treated plants was located opposite to the vectors and distant from the origin of the axes.

All the other pristine MNP-exposed samples grouped together with the control samples, following the vectors of the variables. After the aging process of the particles, the treatment produced more dispersed points in the biplot. aPC samples were the furthest from the origin following the vectors of Zn and Ca ions, while aPE samples were affected by Cu and Fe, albeit to a lesser extent. The other groups, i.e control, aPET and aPVC, were located closer to the vectors of plant length and photosynthetic parameters.

3.6. SEM images

SEM observations showed the typical disposition of *T. usneoides* trichomes on leaves. A central “shield”, strictly adhering to the epidermis, was surrounded by a large and freer wing composed of dead cells (Fig. 9A). The trichomes covered the epidermis (and the stomata underneath) completely and the wings were in many places overlapping (Fig. 9A). The mean dimensions of the trichomes are reported in Table S5. The micromorphology and the dimensions of trichomes of the plants after the exposure to the plastic particles were the same as in control conditions, for both pristine and aged MNP treated samples. In particular, control plants showed trichome dimensions of about $370 \times 235 \mu\text{m}$, with a mean shield diameter around $100 \mu\text{m}$. Even though the treated samples showed, in some cases, different values of the trichomes dimension in respect to control plants, the difference was not significant. In MNP-treated samples, plastic particles were visible on the trichomes, with a preferential allocation on the surface of the wing, rather than on its shield (see for example aPET exposed samples in Fig. 9B). Table S5 reports the distribution of the particles on the shield and on the wing of the samples. Within each treatment, the number of particles present on the wing was significantly higher than that of the shield in all the treated plants, whereas no differences were found among the samples.

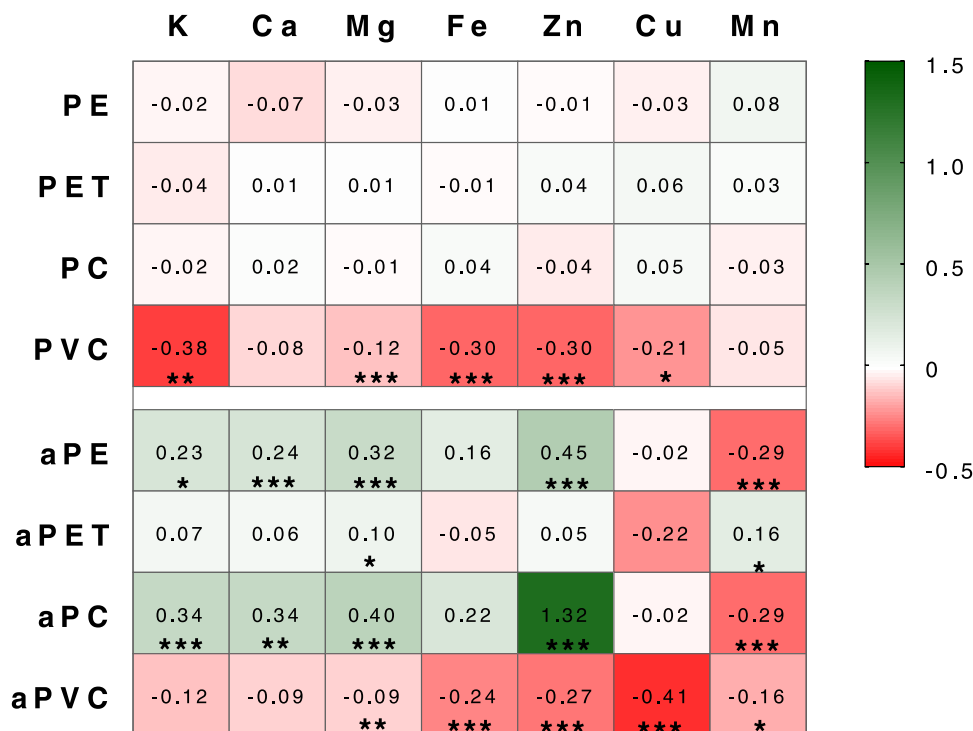
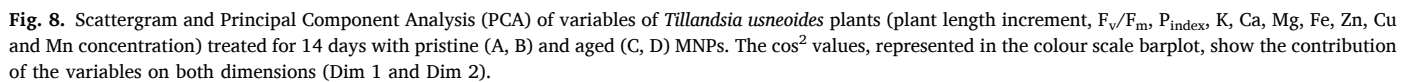


Fig. 7. Heatmaps of ionome variation in strands *Tillandsia usneoides* plants after 14 days of growth in presence of MNPs and aMNPs. Colour scale indicates increased (red), unchanged (white) or decreased (green) element concentration in respect to the corresponding control plants. Asterisks indicate significant differences between treated samples and control samples referred to Table 1 (* = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$).



In this work, an ad hoc setup has been devised to expose *Tillandsia usneoides*, the model plant chosen for monitoring the effect of air contaminants on living organisms, to MNP airborne pollution in controlled conditions. Our results represent the first report showing that airborne pollution at environmentally relevant MNP concentrations can impair plant growth. Previous studies showed that the negative action exerted on plants by MNP pollution in soil and water leads to negative effects on plant diversity and community composition, impacts on ecosystem functions of agroecosystems, and represents a safety risk for the food webs (Rillig et al., 2019; Okeke et al., 2022; Kalčíková, 2020). Here, we demonstrated that also air is among the environmental matrices through which MNPs can represent a threat to plant life, thus posing serious concern about ecological and safety risks for the whole ecosystem. Our result opens the way to further studies aiming to assess the impact of airborne MNP pollution on the shoots of land plants and on the emerged parts of macrophytes, so as to complete the picture on the threat of these new generation of contaminants to plants.

be the most toxic one, almost halving plant length increment at the end of the experiment, and reducing by a third the growth rate. In the case of PE, PET and PC, growth data showed that, after an initial impairment of growth, plants were able to fully adapt to the presence of plastic particles in the air, gaining a final increment in length similar to that of control plants, and a similar growth rate. Thus, pristine PE, PET and PC demonstrated to have minimal toxicity on the studied plant, at least in the adopted experimental conditions. Similarly, other papers studying the impact of soil MNPs reported high toxicity of PVC with respect to other plastic materials, ascribed to the strong ability of this polymer to induce oxidative stress or to misbalance the plant ionome (Pignattelli et al., 2020; Colzi et al., 2022). Indeed, PVC is able to produce long-lived radical species (Rånby et al., 1978) and the highly toxic HCl byproduct (Yuan et al., 2014), which explains its dangerousness. Nonetheless, given the different environmental matrices through which MNPs are administered, a direct comparison with data in the literature is difficult, and MNP specific mechanisms of toxicity on plants might be expected according to their different ways of interaction with air, water and soil.

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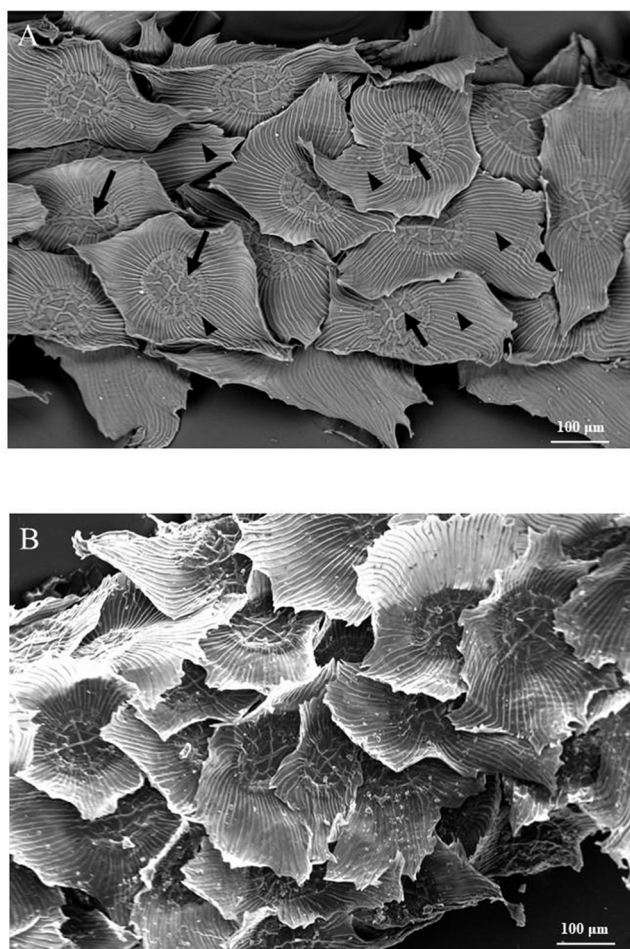


Fig. 9. Representative SEM images of the leaf surface of *Tillandsia usneoides* plants after 14 days of exposure to pristine and aged MNPs. A) Control plants (Mag: 246x): trichomes covering completely the epidermis of the leaf. The trichomes are formed by a central shield (arrows) of 4 cells surrounded by 2 circles of cells and a large external wing (arrowheads) formed by dead cells. The wings overlap one with the other. B) plants exposed to aPET MNPs (Mag: 246x). Many particles (arrows) adhere to the trichome wings and, more rarely, to the trichome shields. (Mag= Magnification).

either increase or decrease the toxicity of such pollutants according to their chemical characteristics. During the two weeks of MNP exposure, significant effects in the reduction of plant length increment appeared at different points in time following the order aPET>aPE>aPC, producing increasing declines in the growth rate. We thus concluded that the aging process made such plastic materials more and differently toxic, with maximum effect for aPC. On the other hand, the more dangerous plastic in its pristine state (PVC) never exerted a significant effect on plant growth after it had been submitted to the weathering treatment. In agreement with ATR evidence of aPVC pronounced oxidation, we can assume that free electrons generated in the pristine polymer were blocked by the newly formed bonds with oxygen atoms, thus leading to a less dangerous material. A similar mechanism has been reported for the case of graphene and graphene oxide (Seabra et al., 2014; Ou et al., 2016; Jia et al., 2019). As for HCl, it is a volatile molecule that can be easily removed by evaporation during the high temperature step of the aging cycle. In any case, also the effect of MNP aging on particle toxicity can be expected to be different according to the matrix where they are present. For example, in the case of soil, opposite results to our study were reported regarding PC impact on *Lepidium sativum*, with the aging process alleviating the contaminant toxicity (Pflugmacher et al., 2021). In that paper, the chemical substances responsible of PC toxicity, such as

Bisphenol A, were supposed to be leached with time during thermal aging. Such reduction of Bisphenol A was found also in our study, nevertheless aPC was able to significantly impair growth starting from the first administration step. Moreover, in a hydroponic experiment the aging of PVC was showed to exacerbate the negative effects of this material on the growth of the alga *Chlamydomonas reinhardtii* (Wang et al., 2020a). As discussed above, a direct comparison with the existing literature is difficult, due to the different experimental conditions (matrix of administration, dimension and concentration of the particles, time of treatment and different kind of aging process). On the other hand, our data contribute to highlight the extreme diversity of MNP behaviour in exerting plant toxicity according to several variables, thus encouraging for further research.

Regarding possible MNP toxicity mechanisms via limitations of photosynthesis, our data proved that airborne plastic pollution could interfere with the photosynthetic process. Leaves of PVC-treated *T. usneoides* showed a relevant reduction of the photosynthetic parameters, with very low values after seven days of exposure and a subsequent partial attenuation of the decrease. Actually, when administered in water and soil, all the studied materials in their pristine state are known to induce reductions in F_v/F_m , P_{index} and other photosynthetic parameters in algae and land plants, even though with mechanisms still unclear and surely multifaceted (see for example Gao et al., 2019; Urbina et al., 2020; Wang et al., 2020a, 2020b; Pflugmacher et al., 2021; Colzi et al., 2022). In the case of airborne pollution, probably the direct interaction with the plant surface was enough to cause photosynthetic damage only in the case of PVC, at least for the experimental conditions here used. Interestingly, the aging of PVC corresponded to a lower negative effect of this plastic on the photosynthetic parameters, that in turn could explain its lower impact on plant growth. The higher toxic effect of the other aged plastic materials was coupled to a greater depression of the photosynthetic process as compared to aPVC, with aPC proving to be the most deleterious pollutant for the photosynthetic activity.

Furthermore, our study demonstrated, for the first time, that the presence of plastic particles in the air can induce significant variations in the plant element concentration, as reported for land plants exposed to MNPs in the soil (Colzi et al., 2022; Fu et al., 2022). Differences in the elemental levels were plastic-specific and varied depending on the aging process (with higher effects for aMNPs, except in the case of PVC) and on the element considered. In any case, such alterations were significant for the plastic materials that impacted more on plant growth, thus can be included in the causes of growth impairment.

In epiphytic rootless bromeliads, like *T. usneoides*, uptake of water and nutrients is achieved exclusively by specialized leaf trichomes (Martin, 1994; Benzing, 2000) with water movement occurring primarily by capillarity among the overlapping trichomes and not by internal transport via the xylem (Herppich et al., 2019). Therefore, since some airborne plastics decreased and some other increased the level of the elements, a possible material-specific interaction of the plastic particles with water droplets present on the plant surface could be hypothesised. Due to the hydrophobic nature and irregular architecture of the plant indumentum, the contact of the shoot surface with water droplets is limited (Pierce et al., 2001). Plastic particles such as aPVC, and even more PVC, adhered to the plant surface could have increased its water repellence, due to the lower wettability of this polymer with respect to the others (as inferred from contact angle values of the studied plastics, <https://www.tstar.com/blog/bid/33845/surface-energy-of-plastics>). Thus, the permanence time of water droplets on the plant surface could have been decreased, resulting in lowered water uptake and element accumulation. The wide PVC-mediated decrease in K concentration could have determined the strong impact of this material on plant growth due to its key role in cell expansion (Marschner, 1995; Ragel et al., 2019). On the other hand, the exposure to PE, PET and PC MNPs in their pristine state seemed to not alter the process of element acquisition, whereas treatment with these materials in their aged state

generally increased the ion concentrations of plants. Since plastic materials have been shown to increase their hydrophilicity upon weathering (Chamas et al., 2020; Chen et al., 2021), the exposure to aMNPs could have lowered the water repellence of the plant surface and increased the contact with droplets, thus increasing water uptake and nutrient accumulation. The excess of elements could have contributed to determine negative effects of aMNPs on growth as well. In any case, direct interactions on the plant surface between the elements present in the water droplets and the particles cannot be excluded and could have led to diverging outcomes, such as in the case of Mn. Probably, this element was bound by some specific aMNPs and made less bioavailable to the plant. Squadrone et al. (2021) showed that microplastics had a relevant role in absorbing and concentrating metals in seawater by several orders of magnitude with respect to the surrounding environment. A similar mechanism could take place in air as well, changing the nutrient balance at the leaf level in *T. usneoides*. In addition, also a direct MNP toxicity on plant development and functioning could occur, further impairing element absorption.

PCA investigation was able to account for a comprehensive behaviour of the studied plant traits under plastic particle treatment and showed that the aging process of the MNPs resulted in a different variation of the affected traits with respect to the pristine materials. Indeed, in the case of pristine MNP treatment, PCA remarkably discriminated PVC-treated samples from all other specimens, which were largely overlapped, suggesting a similar behaviour for most of their traits. The first component of the MNP biplot was mainly characterized by changes in plant growth parameters and no clearly opposite direction of any vectors was scored. Cases did not show a clear location closer to specific traits and were grouped in the directions of the vectors. Only PVC samples were clearly separate showing the opposite behaviour of the other samples. This was in agreement with the evident toxicity of pristine PVC on *T. usneoides* samples, as discussed above. As for aMNP treatment, all the groups were discriminated by PCA, suggesting that, in this case, the exposure to plastic particles affected the traits according to the different material type, whose points were located in distinct regions of the graph. The first component was mainly characterized by the opposite behaviour of plant growth versus Zn, Ca and Mg traits. The aPVC group was the closest to controls, both groups being located near the vectors of plant growth, in agreement with the observed decrease in toxicity of this plastic material when aged. On the other hand, the aPE and aPC cases followed the ion vectors, thus suggesting that the changes in concentrations of these metals was the main factor explaining the growth reduction induced by these aMNPs. The second component showed mostly positive scores of the vectors, with dominance of photosynthetic parameters and Cu and Fe concentration. Plant length was the only trait with negative score, indicating that the growth reduction could be attributed in second instance to changes of the above-mentioned traits, especially for aPE and aPET-treated samples.

Concerning the presence of particles on the strains, SEM images showed that not only MNPs adhered to the plant surface but also had a preferential localization on the trichome wing. This part of the trichome is devoted to take up water and nutrients from the environment (Mosti et al., 2008), with absorption occurring through its dead cells (Brighigna et al., 2002). Even though all the MNPs were shown to adhere to this part, due to their different physico-chemical properties some of them might have interfered with the absorption of water and nutrients, thus contributing to the significant effect exerted on plant growth by certain plastic materials. Similarly, the accumulation of plastic particles on the seed surface has been shown to reduce water absorption in *Lepidium sativum* (Bosker et al., 2019). In addition, since trichomes could be the route of MNP entrance inside the mesophyll, a direct toxic effect of some of these contaminants on plant cells cannot be excluded in explaining their impact on *T. usneoides*.

5. Conclusions

Upon exposing *Tillandsia usneoides* to MNP airborne pollution in controlled conditions, marked depression of plant growth was observed in a polymer type- and aging-dependent manner. PVC was the most toxic material in its pristine state, while PC toxicity was the highest after aging, implying that permanence in the environment can either decrease or increase the impact of plastic particles. MNP toxicity was related to reduction not only of plant growth but also of photosynthetic efficiency, as well as to changes in the micro- and macro-elemental profile. Plastic particles were localised on the leaf trichomes and preferentially on their wings. Our findings are important since they show that airborne plastic pollution can pose direct threat to plants, thus raising concerns about possible negative consequences for the whole ecosystem. Further research should extend this study to other plant species, including crops, in order to evaluate MNP transfer into the food chain and the consequent impact on food quality and human health.

Statement of environmental implication

The materials studied in our work are four types of what is considered the “next generation” contaminant, that is plastic. We administered them as particles at the micro-nanoscale, which is their most hazardous form for the living organisms, to *Tillandsia usneoides* plants through an experimental set-up ad hoc-devised to simulate airborne pollution. Our work helps address environmental problems since it is the first report on the effect of airborne micro-nanoplastic pollution on plants, thus raising concern on a still unknown aspect of the diffusion of such contaminants in the environment and claiming attention for its attenuation.

CRediT authorship contribution statement

Sara Falsini: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – review & editing. **Iliaria Colzi:** Conceptualization, Formal analysis, Investigation, Data curation, Writing – review & editing, Supervision. **David Chelazzi:** Formal analysis, Investigation, Writing – review & editing. **Marco Dainelli:** Formal analysis, Investigation, Writing – review & editing. **Silvia Schiff:** Methodology, Formal analysis, Writing – review & editing. **Andrea Coppi:** Resources, Writing – review & editing. **Alessio Papini:** Data curation, Resources, Writing – review & editing. **Cristina Gonnelli:** Conceptualization, Methodology, Resources, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Sandra Ristori:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Cristina Gonnelli reports financial support was provided by University of Florence.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2022.129314](https://doi.org/10.1016/j.jhazmat.2022.129314).

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