Science of the Total Environment 487 (2014) 206–215

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Atmospheric particulate deposition in temperate deciduous forest ecosystems: Interactions with the canopy and nutrient inputs in two beech stands of Northeastern France

Emeline Lequy [a,b], Christophe Calvaruso [a,1], Sébastien Conil [b], Marie-Pierre Turpault [a,*]

[a] INRA, Biogéochimie des Ecosystèmes Forestiers 1138, F-54280 Champenoux, France
[b] ANDRA, Centre Meuse Haute Marne, F-55290 Bure, France

HIGHLIGHTS

- Atmospheric particulate deposition (APD) is not measured in French forests.
- APD could be a source of nutrients for forests based on poor soils.
- The influence of forest canopy on APD is poorly known.
- Nutrient inputs below canopy were measured in APD and dissolved deposition.
- APD is enhanced by the canopy and contributes to the nutrient inputs of forests.

ARTICLE INFO

Article history:
Received 9 January 2014
Received in revised form 4 April 2014
Accepted 6 April 2014
Available online xxxx

Editor: P. Kassomenos

Keywords:
Atmospheric deposition
Mineral dust
Organic matter
Canopy
Nutrient budgets

ABSTRACT

As wood harvests are expected to increase to satisfy the need for bio-energy in Europe, quantifying atmospheric nutrient inputs in forest ecosystems is essential for forest management. Current atmospheric measurements only take into account the <0.45 μm fraction and dry deposition is generally modeled. The aims of this study were to quantify atmospheric particulate deposition (APD), the >0.45 μm fraction of atmospheric deposition, below the canopy, to study the influence of the canopy on APD, and to determine the influence of APD below canopy to nutrient input–output budgets with a focus on base cations calcium, magnesium and potassium, and phosphorus. APD was sampled every four weeks by passive collectors. We divided APD into an organic and a mineral fraction, respectively POM and MDD. MDD was divided into a soluble and a hardly soluble fraction in hydrogen peroxide, referred to as S-MDD and H-MDD, respectively.

In order to better understand the influence of the canopy on APD, we studied APD in three pathways below the canopy (litterfall, stemflow and throughfall), and in open field. Our results indicated that APD in throughfall (123 ± 64 kg ha^{-1} year^{-1}) was significantly higher and synchronic with that in open field (33 ± 9 kg ha^{-1} year^{-1}) in the two study sites. This concerned both POM and MDD, suggesting a large interception of APD by foliar surfaces, which is rapidly washed off by rain within four weeks. Throughfall H-MDD was the main pathway with an average of 16 ± 2 kg ha^{-1} year^{-1}. Stemflow and litterfall were neglected. In one study site, canopy intercepted about 8 kg ha^{-1} year^{-1} of S-MDD. Although base cations and phosphorus inputs by APD are lower than those of <0.45 μm deposition, they contributed from 5 to 32% to atmospheric deposition and improved the nutrient budget in one of the study sites.

1. Introduction

The European Commission targeted a 20% share of energy from renewable sources (European Convention, 2010). To satisfy this policy, wood harvests are expected to increase by about 30% by 2020 as compared to 2010 (European Convention, 2013). Yet European temperate forests generally grow on acidic and poor-nutrient soils, and fertilizers are rarely used. While carbon and nitrogen cycles have been abundantly documented in forest ecosystems, those of other limiting nutrients such as calcium, magnesium, potassium, and phosphorus still need investigation. Atmospheric deposition is their main external source of these nutrients, backed up with generally lower inputs from soil weathering. Increasing wood harvests might endanger forest sustainability by

* Corresponding author.
E-mail address: turpault@nancy.inra.fr (M.-P. Turpault).
[1] Present address: EcoSustain company, 31 route de Volmerange, F-57330 Kanfen, France.

http://dx.doi.org/10.1016/j.scitotenv.2014.04.028
0048-9697/

causing too large nutrient losses. Negative nutrient budgets in European forests have already been pointed out (Fichter et al., 1998; Ranger and Turpault, 1999; Merino et al., 2005). In this context, it is essential to keep wood production sustainable and preserve the nutrient status of nutrient poor forest soils. This can be done by thoroughly describing nutrient circulation in forest ecosystems and by determining the level of crop harvesting to meet the wood demand, while still keeping the nutrient budgets balanced (Ranger and Turpault, 1999).

In particular, quantifying atmospheric base cations and P inputs is a major stake. So far, forest monitoring networks (e.g. RENECOFOR in France) have quantified atmospheric deposition by analyzing rainwater samples after filtration on 0.45 μm porosity membranes (Ulrich et al., 1998) and modeling dry deposition from throughfall, stemflow and openfield filtrated solutions (Ulrich, 1983). This excludes the >0.45 fraction of atmospheric deposition, named atmospheric particulate deposition (APD). Recently, APD was estimated to enhance base cations and P budgets in forest ecosystems (Lequy et al., 2012). APD was demonstrated to contribute to the nutrition of Holm oak in Northern Spain (Avila et al., 1998), and in a tropical forest (Pett-Ridge, 2009). Many studies investigated the interactions between canopy and aerosols containing nitrogen, sulfur, ozone (Lovett, 1994) and base cations (Draaijers et al., 1997a, 1997b) but the interactions between APD and canopy remain unclear.

APD comprises particulate organic matter (POM) and mineral dust deposition (MDD). MDD in Northeastern France is thought to mainly come from human activities, agriculture and construction works for example, and in a very smaller extent, from large sources of dust, such as the Saharan desert. APD comprises (i) wet deposition, the scavenging of atmosphere by precipitation, (ii) dry deposition, physical processes in the absence of rain, and (iii) fog deposition, the transfer of matter contained in droplets of fog on a solid surface. According to recent models and measurements (Mahowald et al., 2005; Lequy et al., 2013a), mineral dust deposits between 10 and 20 kg ha^{-1} $year^{-1}$ in open field in the region of the present study.

Atmospheric inputs strongly depend on the canopy that intercepts dry and fog depositions. This dry deposition induced by the canopy is currently taken into account in nutrient budgets by modeling. Dry deposition involves complex processes, especially on foliar surfaces, which are now better explained by various mechanistic models and measurements (Lovett, 1994; Marques et al., 2001), as reviewed by Petroff et al. (2008). These models and measurements used wind-tunnels (Reinap et al., 2009) or artificial foliages of needles (Dambrine et al., 1998; Stachurski and Zimka, 2000). They described how leaves of deciduous trees can trap atmospheric components, although their efficiency is lower than that of needles for N and S (De Schrijver et al., 2007). The structure and roughness of the forest canopy resulting from the multiple stages of leaves cause turbulences at the forest boundary layer and increase the particle impaction on the canopy compared to plain surface (Lovett, 1994; Rodrigo and Avila, 2002; Erisman and Draaijers, 2003). Canopy was proven to greatly influence dry deposition in forest ecosystems (Draaijers et al., 1997b; Berger and Glatzel, 1998; De Schrijver et al., 2007). Analyzing nutrient inputs and calculating nutrient budgets in forest ecosystems are hampered by the internal recycling of nutrients, especially by canopy exchange. Canopy exchange comprises the absorption and release of nutrients by the leaves. It disturbs the analyses of the dry deposition induced by canopy and is obviated by Na-based modeling. Na was chosen because it is considered as not affected by canopy exchange (Ulrich, 1983).

In this study, we hypothesized that canopy also enhances APD, and we assumed three pathways of APD below canopy: in throughfall, in stemflow, and by litterfall, as particles may remain trapped on the leaves (Fig. 1).

This study aims at (i) quantifying the 3 pathways of APD below the canopy, (ii) studying the influence of the canopy on APD, and (iii) determining the influence of APD below canopy to nutrient input–output budgets.

For that purpose, APD was measured in open field and below the canopy by typical collectors used in forest monitoring studies and networks in two beech stands on slightly acidic soils of Northeastern France. The deposition rates of APD and its organo-mineral distribution were analyzed and compared in open field and below canopy so as to evaluate the canopy interception of APD. The inputs of APD and their impacts on biogeochemical cycles of temperate forest ecosystems were compared with those of <0.45 μm deposition.

2. Experimental section

2.1. Study sites

Two beech stands were chosen in Northeastern France in the Breuil-Chenue and Montiers-sur-Saulx sites. The soils of the Breuil-Chenue and Montiers-sur-Saulx sites are both alocrisols, respectively acidic and slightly acidic, developed on granite and Cretaceous acid detrital sediments (Table 1).

2.2. Conceptual models of the organo-mineral distribution in APD

In this study, APD in open field is considered as ruled by vertical fluxes in the absence of obstacles for horizontal fluxes. Below the canopy, the >0.45 μm particles transported by lateral winds may be intercepted by the canopy (Fig. 1).

According to the following conceptual model, APD is divided into an organic and a mineral fraction, referred to as POM and MDD, respectively. MDD is composed of easily soluble minerals such as carbonates, and of hardly soluble minerals such as silicates (Lequy et al., 2013a, 2014):

$$APDo = POMo + MDDo = POMo + S{-}MDDo + H{-}MDDo \quad (1)$$

where POMo is the particulate organic matter and MDDo is the mineral dust deposition in open field, S-MDDo is the soluble part of MDDo and H-MDDo is the hardly soluble part of MDDo.

APD below the canopy, APDbc, is made of APD in throughfall (APDt), stemflow (APDsf) and litterfall (APDlf). Eq. (1) is completed by hypothetically supplementary MDD and POM from the interception of the canopy and the litterfall:

$$APDbc = APDt + APDsf + APDlf = APDo + APDc{-}FOM \quad (2)$$

and

$$APDc = POMc + MDDc = POMc + S{-}MDDc + H{-}MDDc \quad (3)$$

where MDDc is the intercepted MDD by the canopy, with a soluble and an insoluble parts, respectively S-MDDc and H-MDDc, POMc is the intercepted POM by the canopy, and FOM is made of internal recycling of matter from the tree tissues and must be subtracted. Similarly, APDsf = POMsf + MDDsf with a soluble and an insoluble fractions.

2.3. Sampling and preparation of the samples

2.3.1. Sampling

Particulate deposition was sampled in open field, in throughfall, stemflow, fresh leaves and litterfall during the April 2010–March 2012 period. In the two sites, a plot of 19 × 19 m was identified in the forest, in which tree diameters at breast height were inventoried and divided into 5 classes. One tree was chosen in each five classes of diameters at breast height to be equipped with the throughfall and stemflow samplers.

The collectors are similar in open field and throughfall. A collecting surface is connected to a 20 L collection polyethylene bottle with a nylon sieve to limit coarse pollution of insects, twigs and leaves. Open field collecting surfaces were polyethylene funnels of 0.22 m^2 with a rim to prevent projection of APD out of the collector. Throughfall collecting surfaces were polyethylene gutters, as those used in other

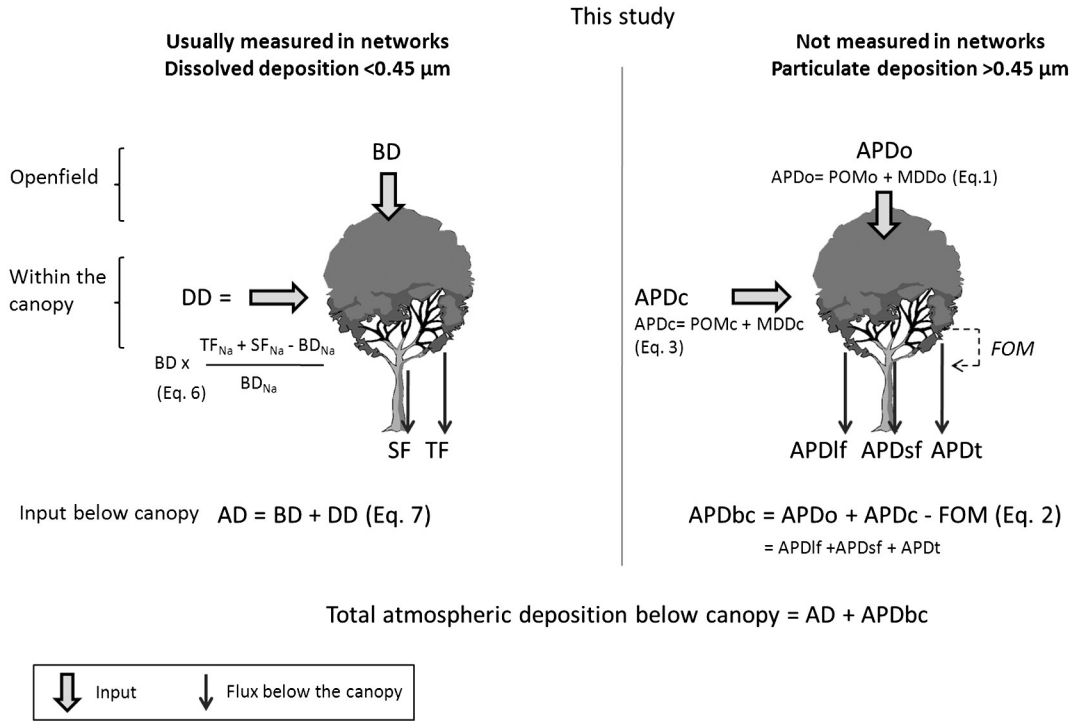

Fig. 1. Fluxes involved in atmospheric dissolved deposition (AD, <0.45 µm) as classically measured and modeled in forest monitoring networks and in this study (Ulrich, 1983; Ulrich et al., 1998), and atmospheric particulate deposition (APD, >0.45 µm) in forest ecosystems specifically measured in this study. BD, DD, and SF are bulk deposition, dry deposition, and stemflow, respectively. MDD, POM, and FOM are the mineral dust deposition, the particulate organic matter of exogenous sources, and the forested organic matter, respectively. The letters bc, o, c, t, sf and lf refer to below canopy, open field, canopy, throughfall, stemflow and litterfall.

forest measurement networks to integrate the variation of foliar density of the canopy because they are placed radially under the canopy.

Stemflow was sampled by a collar of polyurethane foam coated with silicon and directly stuck on the trunk that conveys stemflow by silicon tubing to a 120 L polyethylene container.

Sampling in open field and throughfall followed the same protocol and was described in Lequy et al. (2014). Briefly, the 20 L bottles were siphoned and the collecting surfaces were cleaned with deionized water and brought back to lab. Stemflow containers were equipped of a tap to empty the upper 115 L and the last 5 L were then transferred to the lab after cleaning the 120 L containers.

As the sample preparation was time-demanding, we decided not to treat all the samples individually. In order to study the variations between the collectors, we treated individually the samples from the Montiers-sur-Saulx site, and we pooled the samples from the Breuil-Chenue site, every 4-week periods during all the study period (Table 2). The Montiers-sur-Saulx samples were also pooled during the December 2010–March 2011 and December 2011–March 2012 periods, during which H-MDD was low.

Fresh leaves were gunshot in July 2010 in the Montiers-sur-Saulx site. They were sampled by dry weather to prevent the leaves from a rain-induced wash off.

In the two sites, litterfall was sampled by 5 litter bags with a surface of 1 m^2. They consisted of an open cube of plastic canvas hold 40 cm above the ground by metal stakes.

2.3.2. Sample preparation

The influence of the canopy on APD was analyzed below the canopy in the organic and mineral fractions of APD, according to Eqs. (1) and (2). For this purpose, the samples were prepared with two protocols (Table 2). Thus the samples were divided into 2 sub-samples (Lequy et al., 2014). The "APD" protocol was designed to measure and analyze POM and MDD. The "H-MDD" protocol was designed to separate and study H-MDD, which is a quite constant composition set of hardly soluble crystallized minerals (Lequy et al., 2013a). In the end, the samples were compared (i) between sites in open field and (ii) between open field and below canopy for both sites.

The two protocols started identically by siphoning the samples in field and bringing them back in laboratory. Then the collecting bottles were divided into two sub-samples. They were cleaned with deionized water in centrifugation flasks. Then, the samples were centrifuged at 3500 RPM, transferred in borosilicate beakers after the supernatant was siphoned. The "APD" samples were then oven-dried at 35 °C, weighed and analyzed. The "H-MDD" samples were placed on heating plates at 35 °C with an addition of 8.10^{-5} mol H_2O_2 three times a week until removal of POM. Then the samples were centrifuged at 3500 RPM, the supernatant was removed and the H-MDD samples were oven-dried at 35 °C and weighed. The yield of this protocol was of 69 ± 5%. The APD protocol was applied between September 2011 and March 2012, and the H-MDD protocol was applied between April 2010 and March 2012.

Table 1
Main characteristics of the Breuil-Chenue and Montiers-sur-Saulx sites.

Site	Type of forest		Soil and bedrock		
	Dominant species	Age in 2011 (years)	Type	pH	Bedrock
Breuil-Chenue	*Fagus sylvatica* L.	35	Alocrisol	3.9	Granite
Montiers-sur-Saulx	*Fagus sylvatica* L.	50	Alocrisol	5.1	Acid detritic sediments

Table 2
Organization of the samplers in the Breuil-Chenue and Montiers-sur-Saulx sites. The pathways are open field, throughfall and stemflow.

		Montiers-sur-Saulx	Breuil-Chenue
Study periods	H-MDD	April 2010–March 2012, sampled every four weeks	
	Pooled samples	4-week periods during December 2010–March 2011 and December 2011–March 2012, by pathway	4-week periods, by pathway
	APD	September 2011–March 2012, sampled every four weeks, pooled by month and pathway	
	Fresh leaves	July 2010	No sampling
	Litterfall	After leaf fall	
	Nutrients in S-MDD + POM	April 2011–March 2012	
	Dissolved deposition	January 2011–December 2011, every four weeks	
Number of collectors by pathway	Openfield	4 (2 APD + 2 H-MDD)	
	Throughfall	5 (2 APD + 3 H-MDD)	
	Stemflow	5 (2 APD + 3 H-MDD)	
Number of samples	H-MDD	One sample by collector and 4-week period (except during December 2010–March 2011 and December 2011–March 2012)	One sample by pathway and by 4-week period
	APD	One sample by pathway and by 4-week period	

2.4. Analysis

2.4.1. Quantifying the C&N contents in APD

The samples were then analyzed for their contents in C and N with a CHN «Thermo Quest» Type NCS 2500 (measurement uncertainty less than 5%).

2.4.2. Quantifying chemical contents in MDD

Samples of H-MDD were analyzed by ICP-MS after alkaline fusion in $LiBO_2$ and in HNO_3 at the SARM laboratory (CNRS, Vandoeuvre-lès-Nancy, France). The solution of POM removal was analyzed by ICP-AES. The elements analyzed were Ca, K, Mg, Na and P. Details are available in Lequy et al. (2014).

2.4.3. Analyses of fresh beech leaves and litterfall

Gunshot fresh leaves and litterfall leaves were directly observed by SEM-EDS. Litterfall leaves were washed out with ultrasounds in deionized water. The result was mounted on a glass for X-ray diffraction (XRD) analyses. XRD analyses were performed using a Siemens D5000 diffractometer, equipped with a graphite monochromator and a Cu tube (Cu-Kα radiation). The samples were analyzed within the 1.5 to 70°2-theta range with the following set up: a 0.01°2-theta step, a counting time of 3 s per step and a power of 40 kV and 30 mA. Diffractograms were recorded using a DACO-MP recorder associated with a computer using Diffrac AT software (Socabim, Champs-Sur Marne, France). Minerals were identified using their ASTM files included in the software.

2.5. Calculations of deposition rates

To integrate the differences of surface between open field and throughfall collectors, all the results were normalized to 1 m^2.

2.5.1. Calculations of the fraction POM

POM was calculated from the C content of the samples according to the soil sciences formula Organic matter % = 1.72 × organic carbon % (Pribyl, 2010). This factor may overestimate POM because total C comprised not only OM but also elemental C.

As the nature of POMo and POMt differ, using total C as a proxy for POM may bias the results (Lequy et al., 2014).

2.5.2. Calculations in open field and in throughfall

To compare APD in open field and in throughfall, the rates of APD and of H-MDD were calculated on different time scales: (i) annual scale (April 2010–March 2011 and April 2011–March 2012) and (ii) four-week sampling periodic scale (Rc).

Annual rates in g m^{-2} $year^{-1}$ were calculated as the total weight of the samples of the one-year period normalized to 1 m^2.

Continuous rates in a four-week sampling period i, R_{ci}, in g m^{-2} day^{-1}, were calculated for each site between two consecutive dates of sampling according to Eq. (3).

$$R_{ci} = \frac{W_i}{RR \times S_i \times D_i} \quad (4)$$

where W_i is the weight of the sample during the period of sampling, in grams, S_i is the collecting surface in square meters, RR is the recovery rate of the protocol (72% and 69% for APD and H-MDD, respectively) and D_i is the sampling duration in days.

2.5.3. Calculations in stemflow

The stemflow samples were collected directly on trunks. To normalize the APD and H-MDD stemflow to 1 m^2, the mass of the samples was converted to a deposition rate according to the following formulas.

For each sampling period, the quantity of the stemflow sample is converted to a rate expressed in mass per surface as follows:

$$Msfj = T \times (1/S) \times (1/RR) \times M'sfj \quad (5)$$

where Msfj is the quantity of particles during one period on 1 m^2 (in g m^{-2}), T is the proportionality coefficient taken into account the number of trees on the plot (7.2 and 9.2 in B and M, respectively, as there were 5 trees equipped on the 36 and 46 trees for the 19 × 19 m plots, respectively), S is the plot surface (361 m^2), RR is the recovery rate of the protocol (0.69) and M'sfj is the quantity of particles during one period for the five equipped trees (in grams).

2.5.4. Calculations of dry deposition in <0.45 μm

Atmospheric dry deposition of Ca, Mg, K and P was estimated following the method of Ulrich (1983). This Na-based method assumes no interaction of Na in the canopy so that dry deposition of Na is defined as the difference between throughfall plus stemflow and open field deposition. The dry deposition of cation x (Ca^{2+}, K^+ and Mg^{2+}) was calculated as follows:

$$DDx = BDx \times \frac{TF_{Na} + SF_{Na} - BD_{Na}}{BD_{Na}} \quad (6)$$

where DDx and BDx are the dry deposition and bulk deposition of element x, respectively, and TF_{Na}, SF_{Na} and BD_{Na} are the throughfall,

stemflow and bulk deposition of Na, respectively. Atmospheric dissolved deposition (AD) is calculated as follows:

$$AD = BD + DD. \quad (7)$$

The dry deposition of P was calculated as the difference between the throughfall plus stemflow deposition and the bulk deposition.

2.6. Statistics

2.6.1. Comparing the two types of samplers

To compare the results obtained by funnels in open field and gutters in throughfall, 4 gutters were set up from June to November 2011 next to the funnels in open field. The samples were treated with the "H-MDD" protocol. The winter values, where the leaves are fallen from the beech trees, were added to this comparison. On average, gutters sampled 1.3 ± 0.2 times as much as the funnels for the same sampling periods.

APDt and the H-MDDt values are corrected by the factor 1.3 in the rest of the paper.

2.6.2. Analysis of the influence of the canopy on H-MDD and MDD

Linear regressions were performed for the H-MDD and MDD between the samples in open field and in throughfall.

3. Results and discussion

3.1. Quantification of APD below the canopy

3.1.1. Within the canopy and by litterfall

Fresh beech leaves observed by SEM-EDS revealed particles on the foliar surface, preferentially near the hair-like trichomes of the leave (Fig. 2d). These particles were mainly made of Si and Al (Fig. 2a), with even diatom skulls (white-circled on Fig. 2b), such as those observed in H-MDD in open field (Lequy et al., 2013a). Some particles were also rich in Ca (Fig. 2c).

These coarse minerals on the surface of fresh beech leaves confirm the ability of the canopy to intercept atmospheric mineral particles.

SEM observation of dead beech leaves in litterfall (Fig. 3) revealed some scarce minerals on their surface, covering less than 1% of the foliar surface (quantified after one SEM image). This particle covering seemed lower than that observed on fresh beech leaves, suggesting that a large part of the particles observed on fresh leaves were leached from their surface by rainfall when they are on the tree or on the ground. This is congruent with the results of Stoorvogel et al. (1997) in Ivory Coast, who had observed a rapid scavenging of the particles from the leaves in the rain.

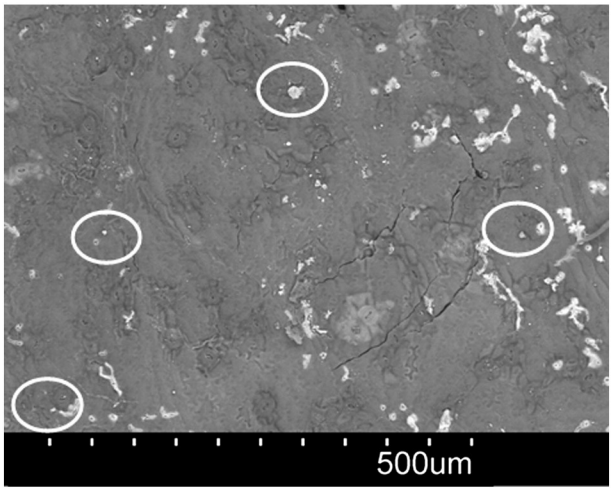

Fig. 3. SEM-EDS observation of the surface of a dead beech leaf collected in a litterbag. Mineral particles are white circled.

The diffractogram of litterfall particles revealed, beyond the organic background, the presence of silicates such as quartz, phyllosilicates and feldspars (Fig. 4). This confirms that the washing off by rain is incomplete and that litterfall constitutes an input of H-MDD for the ecosystem. However, in the absence of any efficient method to extract the mineral particles from the foliar surface, the APD rate by litterfall could not be determined. We assumed that APDlf was negligible as most of the particles are very likely washed off by rain in the throughfall. Eq. (1) is thus rewritten: APDbc = APDt + APDsf.

3.1.2. In stemflow

H-MDDsf was of 0.3 and 0.2 kg ha^{-1} $year^{-1}$ in 2010 and 2011, respectively. This deposition represented about 3% of H-MDD in

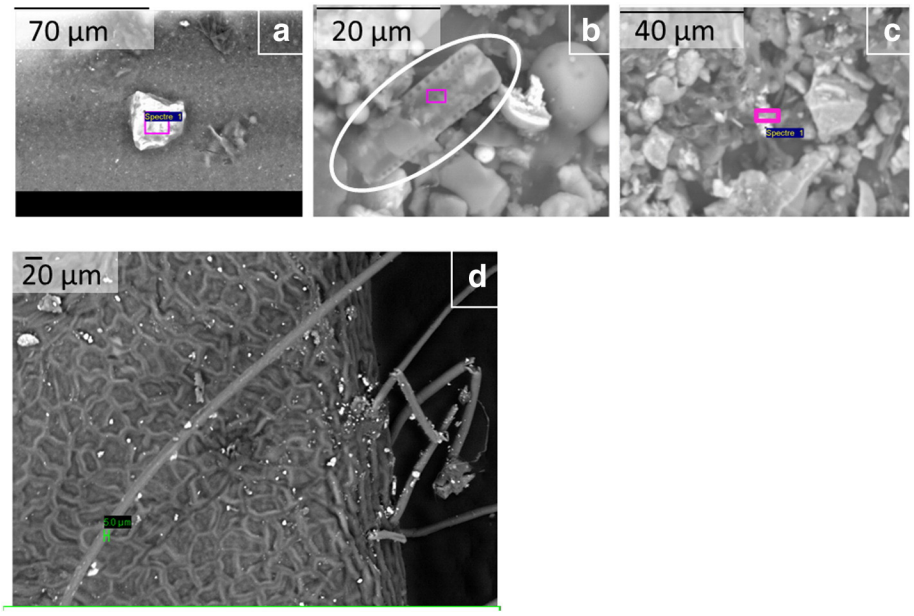

Fig. 2. SEM-EDS photographs of mineral particles observed on fresh beech leaves, dominated by Si (a and b), with a high content of Ca (c), and global view of the edge of a beech leave (d).

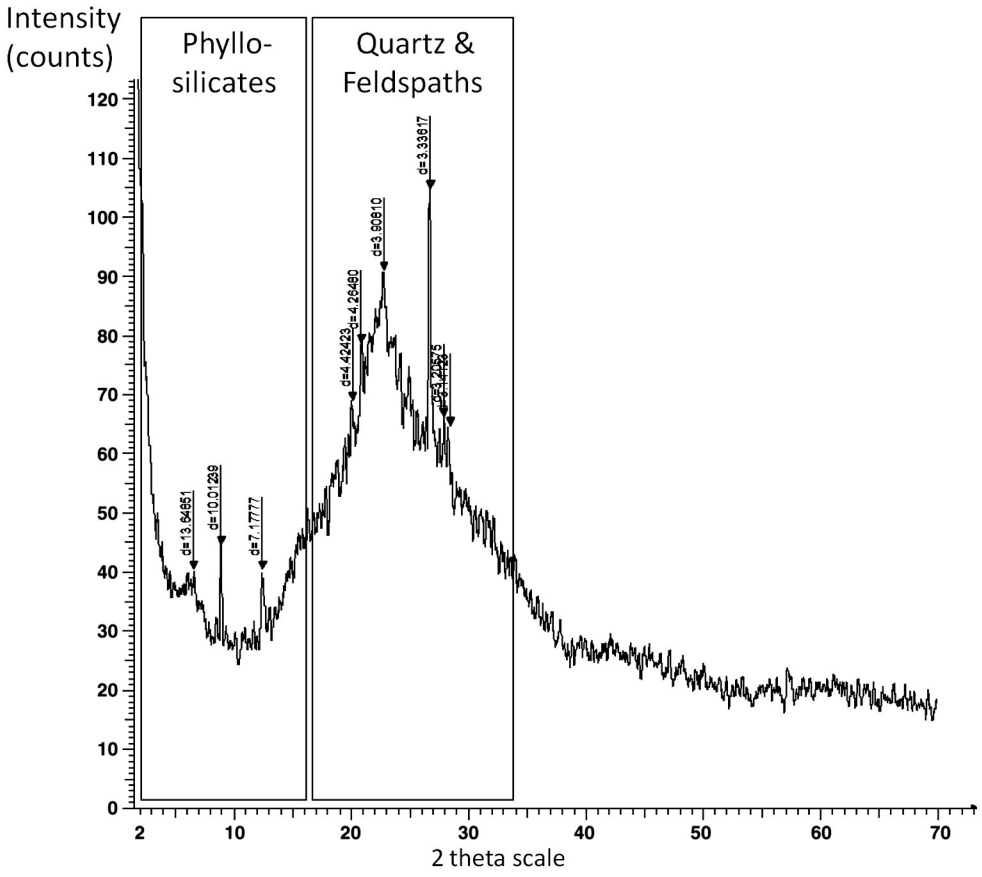

Fig. 4. Diffractogram of the compounds collected on the surface of dead beech leaves in the litterfall of autumn 2010 at the Montiers-sur-Saulx site.

throughfall. H-MDDsf and, by extension, MDDsf were not investigated further, due to a lack of matter for the chemical analysis.

3.1.3. In throughfall

The H-MDD rates in throughfall were similar in the two sites and the two sampling periods with an average of 16 ± 2 kg ha^{-1} $year^{-1}$, which is very similar to the H-MDD rates in open field, 15 ± 3 kg ha^{-1} $year^{-1}$ on average during the same period (Table 3). This is 500 times as high as H-MDDsf. The latter was thus considered as negligible and throughfall was considered as the main pathway of H-MDD below the canopy. The next sections of this paper will focus on APD in throughfall.

In the two sites, the seasonal variations of the average H-MDD rates were quite similar in the 4 collectors in open field and in the 5 collectors in throughfall in 2010 (Fig. 5), with minimum and maximum values in winter, and in spring and summer, respectively. The H-MDD rates were very synchronic in the two sites and did not show any delay with those in open field. This confirms that particles are washed off of the leaves probably within the four-week sampling period and then enrich the H-MDDt. The variability was very high between the samples at the Montiers-sur-Saulx site. As the recovery rate of our protocol was very stable (69 ± 3%), possible explanations would be (i) a heterogeneous particle load of the air during the deposition, (ii) a different deposition of high density particles such as silicates and of lower density such as carbonates between the collectors, or (iii) the variation of the canopy for H-MDDt.

Table 3
H-MDD rates below canopy in litterfall, stemflow and throughfall and in open field in the Breuil-Chenue and Montiers-sur-Saulx sites (kg ha^{-1} an^{-1}) during April 2010–March 2011 (*) and April 2011–March 2012 (**) sampling periods. NA stands for not available. August and September 2011 were not taken into account due to construction works in the Montiers-sur-Saulx site ($^{+}$).

	Breuil-Chenue		Montiers-sur-Saulx	
	2010*	2011**	2010*	2011**,+
Litterfall	–	–	0.1	–
Stemflow	0.3	0.3	0.3	0.2
Throughfall	17.3	16.2	17.6	12.7
Openfield	18.4	12.8	16.2	11.0

3.2. Interception of APD by the canopy

3.2.1. Comparing APD in open field and in throughfall

POM_t was 4.3 and 6.2 times higher than POMo at the Breuil-Chenue and Montiers-sur-Saulx sites, respectively (Fig. 6). The influence of the canopy on POM_t could not be analyzed further. Indeed POM_t is made of three organic sources as $POM_t = POM_o + POM_c + FOM_c$. It was not possible to distinguish $POM_o + POM_c$, that is to say external inputs to the forest ecosystems, from FOM_c, the internal inputs to the forest ecosystem resulting from the interaction of rainfall with twigs and leaves and bringing micro-debris of vegetal tissues in the samples. The C:N ratios in throughfall ranged more widely than those in open field. They reached 27 in throughfall, indicating a contribution of foliar matter as C:N ratios range between 16 and 24 in fresh leaves.

MDDt is 1.7 and 2.6 times higher than in open field at the B and M sites, respectively, suggesting that more MDD was sampled in throughfall during this sampling period.

3.2.2. Comparing H-MDD and S-MDD + POM in open field and in throughfall

This section will focus on H-MDD. Indeed, our dataset for H-MDD sampling is longer than that of APD, which makes possible longer-

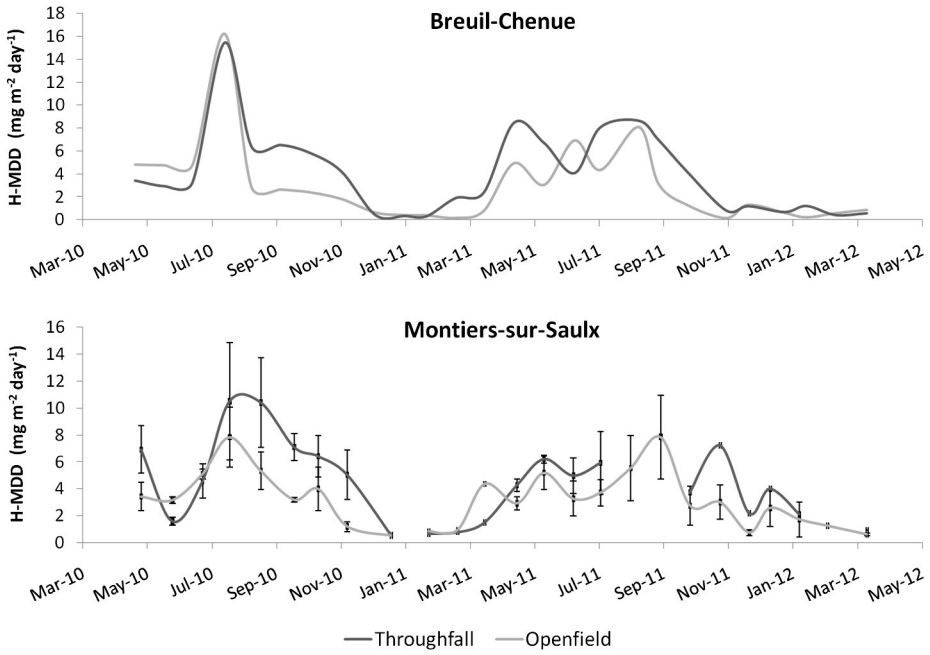

Fig. 5. Average H-MDD rates for 4 collectors in open field (light gray) and for 5 collectors of throughfall in Breuil-Chenue and Montiers-sur-Saulx between April 2010 and March 2012. The bars stand for the standard deviation. All values are in mg m^{-2} day^{-1}.

term comparisons of these two depositions. Besides, H-MDD is the major fraction of MDD as it contributes to 80% of MDD in Northeastern French forests (Lequy et al., 2012).

MDD and H-MDD during the four-week sampling in open field and in throughfall were compared in the Montiers-sur-Saulx site (Fig. 7). H-MDD in open field and throughfall had a linear relationship, H-MDD being slightly higher (about 10%) in throughfall than in open field. MDD in throughfall and in open field had an affine relationship, with a similar slope as that of H-MDD, and a significant intercept at 6.6 mg m^{-2} day^{-1}, i.e. about 15% of the maximal MDD value in throughfall.

However, these results were not statistically significant and highlight the need for further sampling and analysis. This trend yet suggests an interception of MDD by the canopy. Since H-MDD seems to be less influenced by the canopy than the whole MDD, and as MDD = H-MDD + S-MDD, this suggests that the canopy would rather intercept S-MDD (S-MDDc).

If the canopy constantly intercepted 6.6 mg m^{-2} day^{-1} during the 7-month lifetime of canopy every year, this would have provided an additional flux of about 8 kg ha^{-1} $year^{-1}$ of S-MDD in the Montiers-sur-Saulx site.

3.3. Atmospheric nutrient inputs and influence of APD on the biogeochemical cycles

3.3.1. Atmospheric inputs

The APDt inputs in the Montiers-sur-Saulx site were 0.8, 1.1, 0.3 and 0.5 kg ha^{-1} $year^{-1}$ of Ca, K, Mg and P, respectively, and lower than 0.5 kg ha^{-1} $year^{-1}$ for each nutrient in the Breuil-Chenue site (Fig. 8). In the two sites, nutrient depositions are enhanced in APDt compared to APDo, except for Mg and P in the Breuil-Chenue site (Fig. 8). Ca and K contents notably increased in the APD throughfall of the two sites. Their increased content below the canopy can be due to S-MDDc and to FOM, as beech leaves are relatively rich in Ca and K.

Such inputs are similar to those forecasted by the review of Lequy et al. (2012) for MDD in open field in the same area, which estimated a deposition of at least 0.7, 0.4, 0.4 and 0.3 kg ha^{-1} $year^{-1}$ of Ca, K, Mg and P, respectively. The values calculated in open field in the review were probably overestimated, especially those of Ca and Mg, as their atmospheric dissolution in the rain and the collector had not been taken into account in the review (Lequy et al., 2013b). However, the contribution of internal recycling of matter in the ecosystem, FOM, could not be distinguished from that of MDD in our samples.

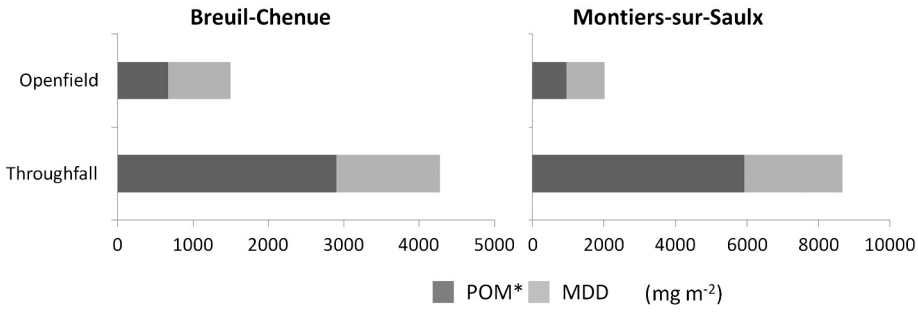

Fig. 6. APD budgets in Breuil-Chenue and Montiers-sur-Saulx in open field and in throughfall between September 2011 and March 2012 (in mg m^{-2}) and its repartition between POM and MDD. *: containing FOM in throughfall.

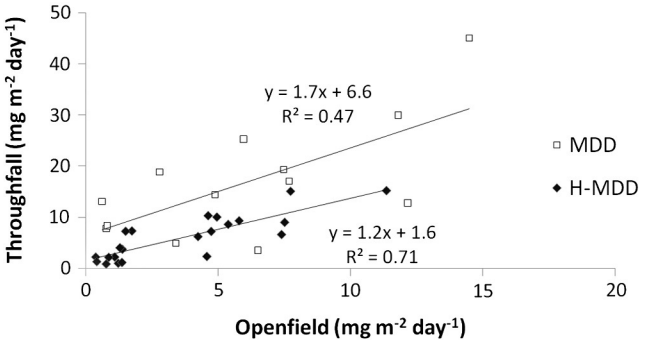

Fig. 7. Daily deposition rates of MDD and H-MDD in throughfall compared to those in open field (mg m^{-2} day^{-1}) at the Montiers-sur-Saulx site.

Results were site- and nutrient-dependent for APDt. Relatively to APDo, the canopy of the Breuil-Chenue site induced an increase of less than 10% of Mg and P and of ca. 50% of particulate Ca and K. The canopy of the Montiers-sur-Saulx site induced an increase of about 25%, 60%, 75% and 90% of particulate Mg, P, K and Ca, respectively. So, in both sites, Mg was the less influenced by canopy, while Ca and K were the most. This indicates a very likely biological contribution (FOM), as base cations abound in the vegetal material (leaves, pollen).

The differences of atmospheric particulate inputs in the two sites may be due to two facts. Firstly, APDo is higher in the Montiers-sur-Saulx site than in the Breuil-Chenue site, leading to a generally higher deposition of nutrients. Indeed, the total atmospheric inputs in the Montiers-sur-Saulx site are from 2 to 7 times higher than those in the Breuil-Chenue site. Secondly, the forest of the Breuil-Chenue site is 15 years younger than that of the Montiers-sur-Saulx site and their management differs. This may cause differences in the canopy structure and explain the differences of the influence of canopy between the two sites.

Large uncertainties remain on the APDt values of this study. By taking into account FOM, which is an internal recycling, we overestimated the nutrient inputs below the canopy. The latter are thus probably between those in APDo and in APDt. The nutrient budgets of this section also lack of consistency with the previous section. Indeed, S-MDD in throughfall was calculated to bring 8 kg ha^{-1} $year^{-1}$ in the Montiers-sur-Saulx site. But, by adding the base cations and P contained in the S-MDD + POM fraction in throughfall, this fraction only brings about 2 kg ha^{-1} $year^{-1}$. This indicates that either we underestimate the fraction S-MDD + POM dissolved in H_2O_2, or our S-MDD calculations via the total APD are overestimated. This inconsistency also may involve particulate nitrogen and sulfur that were not analyzed in the present study. The material used in this study may also lead to underestimation of APD, which could rebound on the reception surface for example. Further investigations are needed on this point.

3.3.2. Comparisons between APD, dissolved deposition and soil weathering flux

Soil weathering was only available for the Breuil-Chenue site (van der Heijden et al., 2013) from the mineralogical data of Mareschal et al. (2012). In the Breuil-Chenue site, the inputs of Ca, K and Mg from APDt reached 10, 40 and 40% of those from bulk deposition, respectively, and 100, 20 and 30% of soil weathering, respectively (Fig. 9). APDbc brought three times as much P as bulk deposition. In the Montiers-sur-Saulx site, the inputs of Ca, K, Mg, and P from APDbc represented about 20, 50, 40 and 60% of those of bulk deposition, respectively.

APDbc provided between 15 and 22% of total atmospheric inputs of Mg and K in both sites, and up to 32% of P in the Breuil-Chenue site. APDbc contributed to a lesser extent on Ca deposition, with 5% and 9% in Breuil-Chenue and Montiers-sur-Saulx, respectively. In the two sites, dry deposition brought at least two times larger inputs of base cations and P than APDbc, and five times and two times larger inputs of P than bulk deposition in the Breuil-Chenue site and the Montiers-sur-Saulx site, respectively. This highlights the importance of dry deposition, as calculated with the method of Ulrich (1983), in the nutrient inputs of forest ecosystems. However, some well-known problems concerning this calculated dry deposition are illustrated in the present study. Dry deposition is calculated from bulk deposition, which contains both wet and dry depositions in open field. Studies carried out with wet-only samples put forward strong differences with bulk deposition (Balestrini et al., 2007; Staelens et al., 2008), and some inaccuracies between the aerodynamic properties of Na-containing aerosols and those for which dry deposition is modeled (Staelens et al., 2008). There is still room for improvement of the canopy budget model, for example (1) in the use of bulk- or wet-only deposition to determine the dry deposition factor, and (2) in the time scale of the model, which should be seasonal and not only annual, so as to analyze the successive phenological periods (Staelens et al., 2008).

For these reasons, even if APDbc was generally lower than the inputs of dissolved deposition (AD) and soil weathering (SW), it cannot be neglected in the two sites. This is especially the case for Ca and P in the Breuil-Chenue site, which represented more than 100% of SW and 300% of AD, respectively. The data about phosphorus is also important, as it may not always be measured in all the studies.

To illustrate the possible influence of APDbc on the nutrition of forest ecosystems based on poor soils, APDbc was finally added to the average input–output budget computed over the 2003–2008 period in the Breuil-Chenue site (van der Heijden et al., 2013) (Table 4). The latter moved towards neutrality of 7%, 20%, 7%, and 10% for Ca, K, Mg, and P, respectively. This forest would still be nutrient deficient according to this budget. However this budget still presents some uncertainties. First of all, the values of APDt for each nutrient are those of the year 2011. We do not know to what extent the values vary over the years. Then, the weathering flux used for the Breuil-Chenue site is an estimation (van der Heijden et al., 2013) and soil weathering is very difficult to

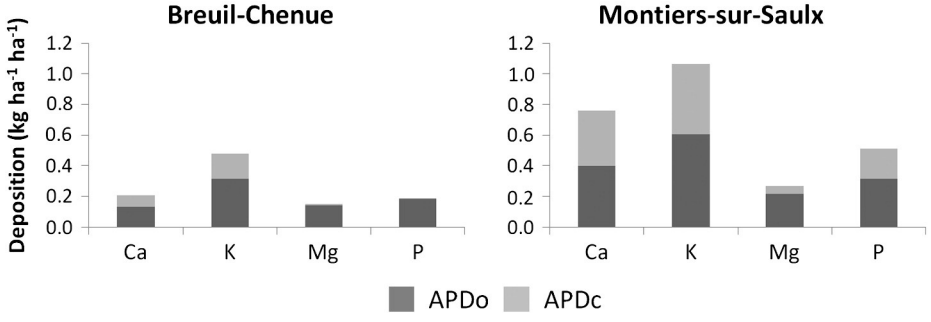

Fig. 8. Annual particulate nutrient inputs in open field (APDo) and intercepted by the canopy (APDc). Their sum is atmospheric particulate deposition below the canopy (APDbc).

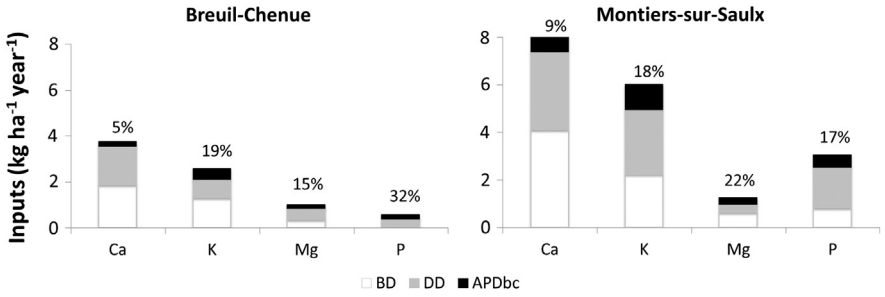

Fig. 9. Atmospheric inputs in the Breuil-Chenue and Montiers-sur-Saulx sampling sites. BD is the dissolved deposition sampled in open field, DD is the dry deposition below canopy calculated according to Ulrich (1983), and APDbc is the atmospheric particulate below the canopy, which still contains the FOM fraction. The percentages are the contribution of APDbc + FOM to atmospheric deposition.

obtain (Klaminder et al., 2011). Finally, dry deposition, as discussed here above, still contains large methodological uncertainties. This budget should be updated in the future with more accurate values.

4. Conclusions

The interest of taking into account APD in nutrient input–output budgets was studied in two beech forests in the North East of France.

Fresh and dead leaves revealed that APD was partly trapped on the surface of leaves within the canopy. It seemed mostly removed from the surface of dead leaves sampled in the litterfall. No delay was noticed between the APD in open field and in throughfall, suggesting that the particles were probably washed off in the next rains to enrich APD in throughfall. Stemflow H-MDD reached 0.3 kg ha^{-1} $year^{-1}$, i.e. less than 3% of throughfall H-MDD. Thus, APD in litterfall and stemflow was neglected below the canopy.

Throughfall is the main contributor of APD below the canopy, as it brought more than 20 kg ha^{-1} $year^{-1}$ of H-MDD. We highlighted that the canopy induced a supplementary POM and MDD fraction to the APD in throughfall. We showed an increase of MDD below the canopy in the mass budget. The additional mineral deposition was mostly S-MDD, which was estimated up to 8 kg ha^{-1} $year^{-1}$. However, we could not clearly separate the organic and mineral origins of base cations and P in APDbc. The external nutrient inputs by APD into forests are probably intermediate between those in open field and those in throughfall.

The enhancement of POM may be due to (1) the increase of the deposition and interception of POM in the canopy and (2) a high fraction of FOM. These two organic compartments are currently not distinguishable.

In forest ecosystems developed on nutrient-poor soils and thriving on small external nutrient inputs, atmospheric particulate deposition is not negligible compared to those of dissolved deposition usually used to assess nutrient budgets. Indeed, it contributed to the total atmospheric input between 5 and 9%, 18 and 19%, 15 and 22%, and 17 and 32%, for Ca, K, Mg and P in the two sites, respectively. This slightly enhanced the nutrient budgets of the Breuil-Chenue site.

As these results came from a one-year study, there is clearly a need for a longer monitoring to clarify the effect of canopy on the nutrient inputs below the canopy and their organic or mineral origin.

Acknowledgments

This study was funded by the Andra. We would like to thank the INRA and the GIP ECOFOR for their technical support and the ONF for letting us access the sites. The BEF unit research is supported by the French National Research Agency through the Laboratory of Excellence ARBRE (ANR-12-LABXARBRE-01).

We are very grateful to Elizabeth Leclerc (ANDRA) for this project, Serge Didier and Jérôme Demaison (INRA) for designing, making and setting up the samplers, to Carine Cochet, Perrine Mathieu, Claire Pantigny, Gregory van der Heijden, Arnaud Legout (INRA) and Maxime Simon (Andra), for sampling and preparing the samples, to Patrick Riveron (INRA) for the analyses of C&N, to Christophe Rose (INRA) for the SEM-EDS and to Pierre Montpied (INRA) for his help with statistics, and the two anonymous reviewers who helped us improve the manuscript.

Table 4
Nutrient budgets without and with APDbc of Ca, Mg, K, and P in the Breuil-Chenue site.

Nutrient budget in the Breuil-Chenue site	Ca	Mg	K	P
Without APDbc	−3.1	−0.8	−7	−1.6
With APDbc	−2.9	−0.6	−6.5	−1.4

References

Avila A, Alarcon M, Queralt I. The chemical composition of dust transported in red rains — its contribution to the biogeochemical cycle of a Holm oak forest in Catalonia (Spain). Atmos Environ 1998;32(2):179–91.
Balestrini R, Arisci S, Brizzio MC, Mosello R, Rogora M, Tagliaferri A. Dry deposition of particles and canopy exchange: comparison of wet, bulk and throughfall deposition at five forest sites in Italy. Atmos Environ 2007;41(4):745–56. [février].
Berger TW, Glatzel G. Canopy leaching, dry deposition, and cycling of calcium in Austrian oak stands as a function of calcium availability and distance from a lime quarry. Can J Forest Res 1998;28(9):1388–97.
Dambrine E, Pollier B, Bonneau M, Ignatova N. Use of artificial trees to assess dry deposition in spruce stands. Atmos Environ 1998;32(10):1817–24.
De Schrijver A, Geudens G, Augusto L, Staelens J, Mertens J, Wuyts K, et al. The effect of forest type on throughfall deposition and seepage flux: a review. Oecologia 2007; 153(3):663–74.
Draaijers GPJ, VanLeeuwen EP, DeJong PGH, Erisman JW. Base cation deposition in Europe part I. Model description, results and uncertainties. Atmos Environ 1997a;31(24):4139–57.
Draaijers GPJ, VanLeeuwen EP, DeJong PGH, Erisman JW. Base-cation deposition in Europe part II. Acid neutralization capacity and contribution to forest nutrition. Atmos Environ 1997b;31(24):4159–68.
Erisman JW, Draaijers GPJ. Deposition to forests in Europe: most important factors influencing dry deposition and models used for generalisation. Environ Pollut 2003; 124(3):379–88.
European Convention. Directive 2009/28/EC of 23 April 2009 on the promotion of the use of energy from renewable sources and amending and subsequently repealing Directives 2001/77/EC and 2003/30/EC; 2010.
European Convention. A new EU Forest Strategy: for forests and the forest-based sector Brussels, 2013 09; 2013.
Fichter J, Dambrine E, Turpault MP, Ranger J. Base cation supply in spruce and beech ecosystems of the Strengbach catchment (Vosges mountains, N–E France). Water Air Soil Pollut 1998;104(1–2):125–48.
Klaminder J, Lucas RW, Futter MN, Bishop KH, Kohler SJ, Egnell G, et al. Silicate mineral weathering rate estimates: are they precise enough to be useful when predicting the recovery of nutrient pools after harvesting? For Ecol Manage 2011;261(1):1–9.
Lequy E, Conil S, Turpault M-P. Impacts of Aeolian dust deposition on European forest sustainability: a review. For Ecol Manage 2012;267:240–52.
Lequy E, Legout A, Conil S, Turpault MP. Aeolian dust deposition rates in Northern French forests and inputs to their biogeochemical cycles. Atmos Environ 2013a;80:281–9.
Lequy E, Nicolas M, Conil S, Turpault M-P. Relationship between atmospheric dissolved deposition and mineral dust deposition in French forests. Water Air Soil Pollut 2013b;224(9):1680.
Lequy E, Conil S, Turpault M-P. Complementary methods to distinguish organic and mineral matter in atmospheric particulate deposition and their respective nutrient inputs to temperate forest ecosystems. Aeolian Res 2014;12:101–9. [Mar].
Lovett GM. Atmospheric deposition of nutrients and pollutants in North-America — an ecological perspective. Ecol Appl 1994;4(4):629–50.

Mahowald NM, Baker AR, Bergametti G, Brooks N, Duce RA, Jickells TD, et al. Atmospheric global dust cycle and iron inputs to the ocean. Global Biogeochem Cycles 2005;19(4). [GB4025].
Mareschal L, Turpault MP, Bonnaud P, Ranger J. Relationship between the weathering of clay minerals and the nitrification rate: a rapid tree species effect. Biogeochemistry 2012;112(1-3):293–309.
Marques MC, Gravenhorst G, Ibrom A. Input of atmospheric particles into forest stands by dry deposition. Water Air Soil Pollut 2001;130(1–4):571–6.
Merino A, Balboa MA, Soalleiro RR, Gonzalez JGA. Nutrient exports under different harvesting regimes in fast-growing forest plantations in southern Europe. For Ecol Manage 2005;207(3):325–39.
Petroff A, Mailliat A, Amielh M, Anselmet F. Aerosol dry deposition on vegetative canopies. Part I: review of present knowledge. Atmos Environ 2008;42(16):3625–53.
Pett-Ridge JC. Contributions of dust to phosphorus cycling in tropical forests of the Luquillo Mountains, Puerto Rico. Biogeochemistry 2009;94(1):63–80.
Pribyl DW. A critical review of the conventional SOC to SOM conversion factor. Geoderma 2010;156(3–4):75–83. [May 15].
Ranger J, Turpault MP. Input–output nutrient budgets as a diagnostic tool for sustainable forest management. For Ecol Manage 1999;122(1–2):139–54.
Reinap A, Wiman BLB, Svenningsson B, Gunnarsson S. Oak leaves as aerosol collectors: relationships with wind velocity and particle size distribution. Experimental results and their implications. Trees-Struct. Funct. 2009;23(6):1263–74.
Rodrigo A, Avila A. Dry deposition to the forest canopy and surrogate surfaces in two Mediterranean holm oak forests in Montseny (NE Spain). Water Air Soil Pollut 2002;136(1–4):269–88.
Stachurski A, Zimka JR. Atmospheric input of elements to forest ecosystems: a method of estimation using artificial foliage placed above rain collectors. Environ Pollut 2000; 110(2):345–56.
Staelens J, Houle D, De Schrijver A, Neirynck J, Verheyen K. Calculating dry deposition and canopy exchange with the canopy budget model: review of assumptions and application to two deciduous forests. Water Air Soil Pollut 2008;191(1–4):149–69.
Stoorvogel JJ, VanBreemen N, Janssen BH. The nutrient input by Harmattan dust to a forest ecosystem in Cote d'Ivoire, Africa. Biogeochemistry 1997;37(2):145–57.
Ulrich B. Interaction of forest canopies with atmospheric constituents: SO2, alkali and earth alkali cations and chloride. Effects of accumulation of air pollutants in forest. Ecosystems 1983:33–45.
Ulrich E, LeLong N, Lanier M, Schneider A. Regional differences in the relation between monthly precipitation and bulk concentration in France (renecofor). Water Air Soil Pollut 1998;102(3–4):239–57.
Van der Heijden G, Legout A, Pollier B, Mareschal L, Turpault M-P, Ranger J, et al. Assessing Mg and Ca depletion from broadleaf forest soils and potential causes — a case study in the Morvan Mountains. For Ecol Manage 2013;293:65–78.