



Complementary methods to distinguish organic and mineral matter in atmospheric particulate deposition and their respective nutrient inputs to temperate forest ecosystems



Émeline Lequy^{a,b}, Sébastien Conil^b, Marie-Pierre Turpault^{a,*}

^aINRA, Biogéochimie des Écosystèmes Forestiers UR 1138, F-54280 Champenoux, France

^bANDRA, Centre Meuse Haute Marne, F-55290 Bure, France

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ABSTRACT

Sampling atmospheric particulate deposition (APD) in forest ecosystems highlights the need for methods to measure and analyze its organic and mineral repartition. We validated an organo-mineral repartition model of APD composition in open fields and below canopy with a mineral fraction, named mineral dust deposition (MDD), and particulate organic matter (POM). MDD is subdivided into soluble (S-MDD) and hardly soluble (H-MDD) fractions. To (i) monitor APD and its nutrient fluxes in forest ecosystems in the north of France and (ii) quantify the relative contribution of POM and MDD to APD, we adapted sampling materials and preparation methods that were developed for regions close to mineral dust sources. We have also compared two protocols. The “APD” protocol led to quick results for APD rates and POM contents. The “H-MDD” protocol is a treatment for soil samples that uses hydrogen peroxide, which solubilized both POM and S-MDD, and allowed detailed analyses of H-MDD. Both protocols induced a mass loss that was a maximum for the “H-MDD” protocol ($31 \pm 3\%$). The contribution of POM in APD in open fields ($49 \pm 10\%$) was lower than below the canopy (at least $66 \pm 6\%$). H-MDD accounted for approximately 80% of the MDD mass and contained the largest portion of low-solubility elements (Si, Al and Fe). The fractions S-MDD and POM contained the largest portion of Ca and P (more than 70%). The two protocols were complementary and may be used successively to accurately describe APD.

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1. Introduction

Great effort has been made in the past few decades to better understand the biogeochemical cycles of major nutrients in forests. These cycles are models that describe the circulation between soil, plants and atmosphere, including the external inputs and outputs of nutrients. Atmospheric particulate deposition (APD) is defined as the $>0.45 \mu\text{m}$ fraction of atmospheric deposition. It is the complementary fraction of the $<0.45 \mu\text{m}$ atmospheric deposition currently sampled in forest monitoring networks. APD holds both organic and mineral portions, respectively referred to as particulate organic matter (POM) and mineral dust deposition (MDD). APD is still not sampled or analyzed in Northern Europe, yet MDD contributes to the nutrition of various ecosystems, bringing nutrients to phytoplankton in the ocean (Mahowald et al., 2005) and to various forest ecosystems (Swap et al., 1992; Avila et al., 1998; Pett-Ridge, 2009). MDD is also thought to contribute to the sustainability of managed forests in acidic soils (Lequy et al.,

2012). The aim of this study is to propose materials and methods suitable for sampling and analyzing APD in European temperate forests.

While carbon and nitrogen cycles have been extensively investigated, those of limiting nutrients such as Ca, K, Mg and P are still not completely understood. Studying APD may contribute to the better understanding of these cycles. In particular, studying MDD is very important for the following three reasons:

1. MDD is nutrient rich, as it contains up to 3% Ca in studies conducted in Spain (Avila et al., 1998). It originates in eroded soil from local or remote areas, which is transported in the atmosphere (Field et al., 2009). MDD is made of various nutrient-containing minerals, from those that are readily soluble such as calcite, to minerals that are less soluble such as quartz (Lequy et al., 2012). In comparison, POM is mostly carbonaceous, as it is composed of pollens, spores, primary and secondary aerosols (Jacobson et al., 2000) and most likely organic matter (OM) associated with eroded soil particles or with anthropogenic aerosols. Analyzing the chemical composition of the entire APD would not allow for distinguishing between POM and MDD elements.

* Corresponding author. Tel.: +33 3 83 39 40 75.

E-mail address: turpault@nancy.inra.fr (M.-P. Turpault).

- The nutrient release from MDD depends on its mineralogical composition, which is impossible to determine by X-ray diffraction in the presence of POM because POM is not crystallized. In the atmosphere, $>1 \mu\text{m}$ aerosols from dust plumes close to emission sources are composed of 13% POM and 1% elemental carbon (EC), while those in Northwestern Europe are composed of 27% POM and 2% EC (Putaud et al., 2004). This suggests a large contribution of POM to APD and therefore a very difficult determination of the mineralogical composition of APD. Additionally, measurements of $<0.45 \mu\text{m}$ deposition indicate a strong contribution of internal recycling of elements below the forest canopy (Ulrich, 1983). This suggests increased POM below canopy, which must be tested. Studying the effect of the canopy on MDD and POM would be impossible without studying them separately.
- APD studies are quite scarce, and POM treatments vary considerably between studies (Stoorvogel et al., 1997; Avila et al., 1998; Kufmann, 2006). It is therefore impossible to compare these measures when POM is included, as they vary depending on spatio-temporal parameters. For example, near the Sahara Desert, which is the largest source of particles in the world (Goudie and Middleton, 2006), MDD clearly dominates the sample. Such is not the case in Northern Europe, at a distance of at least 500 km from the Sahara Desert, where the fraction of POM was higher (Putaud et al., 2004). Quantifying APD and its organo-mineral repartition is important and will also provide data about POM in open fields and in forest.

As the dissolution rates of minerals contained in MDD range from very low (quartz) to very high (calcite), we suggest separating the readily soluble and less-soluble minerals by setting a solubilization limit. This limit will depend on the treatment applied to the samples. We propose the following description of the organic and mineral fractions of the APD in open fields, as MDD associated with POM Eq. (1):

$$\text{APD}_o = \text{POM}_o + \text{S-MDD}_o + \text{H-MDD}_o \quad (1)$$

where POM_o is the atmospheric particulate organic matter, S-MDD_o and H-MDD_o are the soluble and the hardly soluble fractions of MDD, respectively, and o stands for open fields.

The hypotheses to test are summarized in the conceptual model (Fig. 1).

Below the canopy, the interaction of the APD with foliage would cause additional inputs of POM and MDD (Ulrich, 1983; Nemitz et al., 2002; Reinap et al., 2009). Moreover, in forest ecosystems, supplementary organic matter is mixed in the samples below the canopy, where leaf and bark wood abrasion by rain produces

vegetal microdebris that contaminates the APD sampling. Therefore, we hypothesized that POM would strongly contribute to APD in throughfall (APD_t) and proposed to complete Eq. (1) for samples below the canopy as follows:

$$\begin{aligned} \text{APD}_t &= \text{APD}_o + \text{APD}_c \\ &= \text{APD}_o + \text{POM}_c + \text{FOM} + \text{S-MDD}_c + \text{H-MDD}_c, \end{aligned} \quad (2)$$

where APD_o is in open fields samples, POM_c , S-MDD_c and H-MDD_c are the hypothetical supplements added by the canopy, and FOM is the organic fraction coming from the abrasion of twigs and leaves by rain. The letters t and c stand for throughfall and canopy, respectively.

Sampling APD requires passive collectors to sample bulk deposition, either during rainfall or in the absence of rain. Such collectors are based on a common simple principle of a collecting funnel connected to a bottle container.

Sampling and preparing APD varied considerably between studies. Near the Sahara Desert, the coarse debris of POM are often removed after visual observation, as seen in Drees et al. (1993) and in Loye-Pilot and Martin (1996). APD is collected after filtration on 0.40 or 0.45 μm porosity membranes (Loye-Pilot and Martin, 1996; Avila et al., 1997; Stoorvogel et al., 1997) and weighed. Stoorvogel and coauthors (1997) corrected the results obtained from the raw samples to estimate MDD by using the element Ti. In Europe, two studies in Germany (Kufmann, 2006) and in Northern Finland and Sweden (Franzen et al., 1994) sampled APD on a snowy surface. Both studies noted a large share of POM in their samples, 44% and 30% in Germany and in Northern Finland and Sweden, respectively, measured after the removal of POM with H_2O_2 and burning at 700 °C. The measures of Franzen and coauthors (1994) focused on an extraordinary Saharan outbreak, whose mineral fraction might have strongly contributed to APD. The German sampling lasted a year and a half and is likely more representative of APD in general and POM in particular. These two studies highlighted the extent to which POM contributed to open fields APD, but they did not provide any indication of POM and MDD contribution to APD inside forest ecosystems.

This study marks the first time that APD will be sampled in European temperate forests and treated with the standard soil chemical protocol, using hydrogen peroxide to exclude POM from the samples. Our study compared the two protocols. The “APD” protocol keeps APD intact, while the other solubilizes POM in warm hydrogen peroxide. These two protocols were applied on APD samples in open fields and below the canopy in two forest stands. They are described in section 2 and discussed in section 3. The pros and cons of each method are explored, which depend on whether the focus of the study is APD or MDD.

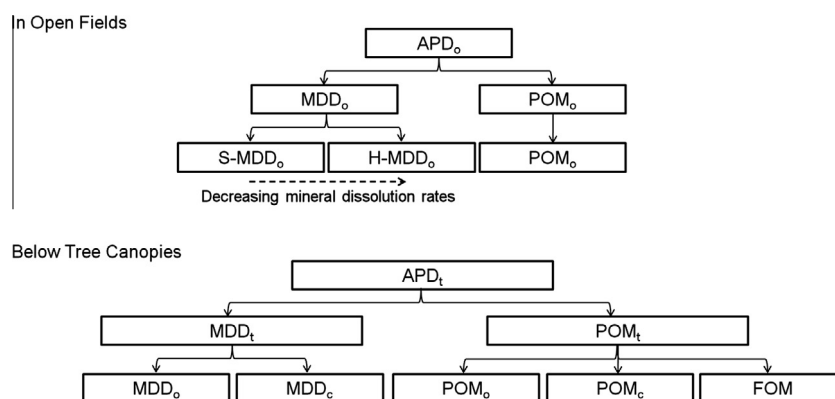


Fig. 1. Schematic representation of the hypothesized composition of APD in this study.

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