Ecological Indicators 79 (2017) 382–390

Contents lists available at ScienceDirect

Ecological Indicators

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

Original Articles

Leaf litter decomposition as a bioassessment tool of acidification effects in streams: Evidence from a field study and meta-analysis

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ARTICLE INFO

Keywords:
Atmospheric acid deposition
Functional integrity
Geology
Litter identity
Mesh size
pH

ABSTRACT

Atmospheric acid deposition affects many streams worldwide, leading to decreases in pH and in base cations concentrations and increases in aluminum (Al) concentration. These changes in water chemistry induce profound changes in the diversity, structure and activity of biological communities and in ecosystem processes. However, monitoring programs rely only on chemical and structural indicators to assess stream integrity. Nevertheless, the ability of ecosystems to provide services rely on their functional integrity and thus ecosystem processes should be considered in monitoring programs. We assessed the potential for leaf litter decomposition, a fundamental ecosystem process in forest streams, to be used as a bioassessment tool of acidification effects on stream ecosystem functioning. In a field study in the Vosges Mountains (North-eastern France), using three leaf litter species (*Alnus glutinosa*, *Acer pesudoplatanus* and *Fagus sylvatica*) enclosed in fine and coarse mesh bags and incubated in streams flowing over granite or sandstone bedrock along an acidification gradient, we assessed if the response of litter decomposition to acidification depended on litter species, mesh size, parent lithology and acidification level. In a meta-analysis of 17 primary studies on the effect of acidification on leaf litter decomposition, reporting 67 acidified – reference stream comparisons, we assessed the consistency in the response of litter decomposition to acidification cross studies and the robustness of litter decomposition to be used as a bioassessment tool. Both the field study and meta-analysis revealed an overall strong inhibition (> 60%) of leaf litter decomposition in acidified streams likely resulting from previously well described altered decomposer community structure and activity. No effect of leaf species was found in the field study, while in the meta-analysis inhibition of leaf litter decomposition in acidified streams was stronger for *Fagus* than for *Acer*, *Quercus* and *Liriodendron*. However, differences among leaf species in the meta-analysis might have been confounded by other differences among studies. The response of leaf litter decomposition to acidification was stronger in coarse than in fine mesh bags, indicating strong impairment of detritivore community structure and activity. The magnitude of inhibition also depended on parent lithology, but this is likely related to differences in the degree of acidification. Indeed, the magnitude of the inhibition of leaf litter decomposition increases with increases in H^+ in Al concentration. Litter decomposition has the potential to be used as a bioassessment tool of acidification effects in streams since it shows consistent response to acidification across regions and is robust to experimental choices.

1. Introduction

Atmospheric acid deposition has drastically affected terrestrial and aquatic ecosystems over large temperate areas of the northern hemisphere (Driscoll et al., 2001) and it is an important emerging problem in Asia (Lu et al., 2010; Liu et al., 2011). The unanimous acknowledgment of the deleterious impacts of atmospheric acid deposition on ecosystems led to the implementation of several national and international rigorous agreements aiming at reducing transboundary air pollution (Likens et al., 2001). Recent decades have indeed witnessed a large decrease in the emission of pollutants and in turn in acid deposition in North America and Europe (Waldner et al., 2014; Lawrence et al., 2015). However, the decrease in acid deposition is not always translated into improved water quality because (i) sulfur compounds accumulated over decades of SO_2 atmospheric deposition are still being leached from soils into freshwaters, (ii) there is an increase in NH_3 emissions from

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http://dx.doi.org/10.1016/j.ecolind.2017.04.044
Received 22 January 2017; Received in revised form 1 April 2017; Accepted 4 April 2017
1470-160X/

intensification in agriculture and in cattle production, (iii) base cations in catchments in acid-sensitive regions often continue to be depleted, and (iv) there is a decrease in base cations atmospheric deposition (Likens et al., 1996; Alewell et al., 2001; Evans et al., 2001; Liu et al., 2011). Nevertheless, evidence of chemical recovery has been reported for several areas with stream water showing declining concentrations of sulphate (SO_4) and aluminum (Al) and increasing pH and acid neutralizing capacity (ANC) (Stoddard et al., 1999; Skjelkvale et al., 2005). But, if signs of chemical recovery have been reported, evidence of biological recovery remains rare (Malcolm et al., 2014a,b) and when it occurs changes in communities (e.g., return of acid sensitive species) appear modest (Monteith et al., 2005). Thus, acidification of freshwaters remains an environmental problem and many ecosystems are still severely affected by water that is chronically or episodically acidic.

Environmental quality assessment of streams is generally based on community (e.g., benthic macroinvertebrates, diatoms and fish) structural variables (Birk et al., 2012; O'Brien et al., 2016). However, community structure and ecosystem function are not always closely coupled and several proposals have been made for the incorporation of ecosystem processes in bioassessment programs (Gessner and Chauvet, 2002; Young et al., 2008).

Leaf litter decomposition is a fundamental ecosystem process in forest headwater streams, since primary production is limited by shading and leaf litter of terrestrial origin constitutes the main source of energy and carbon for aquatic communities (Wallace et al., 1997). The rate at which litter decomposes depends on litter intrinsic characteristics, microbial (mainly aquatic hyphomycetes) and invertebrate consumer (i.e., shredders) activity, and environmental conditions (Webster and Benfield, 1986). Generally, soft litter with high nutrient concentration (i.e., high quality litter) decomposes faster than more recalcitrant litter since microbial colonization is faster and microbial activities are higher in the former than in the latter substrate (Gessner and Chauvet, 1994; Gulis et al., 2006). Leaf litter decomposition is also stimulated in the presence of shredders and increases with increases in their density (Taylor and Chauvet, 2014). Changes in environmental conditions can affect litter mass loss directly, and indirectly by altering community structure and activity of microbial and invertebrate decomposers (Webster and Benfield, 1986).

Stream acidification, and associated increase in monomeric Al concentration and decrease in base cations concentrations, generally inhibits leaf litter decomposition rates (Dangles and Guérold, 1998, 2001a,b; Dangles et al., 2004; Baudoin et al., 2008; Simon et al., 2009; Cornut et al., 2012). This is achieved via inhibition of microbial activities (Griffith et al., 1995; Dangles et al., 2004; Simon et al., 2009), reduction of microbial biomass (Griffith and Perry, 1994; Meegan et al., 1996; Dangles et al., 2004), and reduction of aquatic hyphomycete species richness (Baudoin et al., 2008; Cornut et al., 2012). Also, there is disappearance of acid-sensitive detritivores such as gammarids, sericostomatids and limnephilids that are also large and/or efficient shredders, and reduction of shredder biomass (Meegan et al., 1996; Dangles and Guérold, 1998, 2001a,b; Dangles et al., 2004; Simon et al., 2009). Thus, leaf litter decomposition is particularly interesting as a potential bioassessment tool for addressing acidification effects on stream functioning since it is a key ecosystem process, which has been widely studied and whose response to acidification can be predicted a priori (Dangles et al., 2004; Young et al., 2008; Simon et al., 2009).

In this study, we assessed the potential for leaf litter decomposition to be used as a bioassessment tool to detect acidification effects on stream ecosystem functioning. By performing a field experiment where three leaf litter species were enclosed in fine and coarse mesh bags and incubated in streams flowing over granite or sandstone bedrock along an acidification gradient, we assessed if the response of leaf litter decomposition to acidification depended on litter species, mesh size, parent lithology and acidification level. By performing a meta-analysis of primary studies on the effects of acidification on leaf litter decomposition, we assessed the consistency in the response of leaf litter decomposition to acidification across studies and the robustness with which leaf litter decomposition may be used as a bioassessment tool. We expected a strong inhibition of leaf litter decomposition with increased acidification (i.e., decrease in pH or increase in H^+ concentration) and Al concentration (Dangles et al., 2004; Simon et al., 2009; Cornut et al., 2012; Clivot et al., 2014). This inhibition should be especially strong for soft leaf litter with high nutrient concentration, since microbial activities and shredder contribution to litter mass loss are generally higher in this litter than in more recalcitrant litter (Hieber and Gessner, 2002; Gulis et al., 2006). Since some highly efficient detritivores are acid-sensitive species (Meegan et al., 1996; Dangles and Guérold, 1998, 2001a,b), the inhibition of leaf litter decomposition with acidification should be especially strong for litter incubated in coarse mesh bags.

2. Material and methods

2.1. Field study

2.1.1. Streams

Eight 1st–2nd order streams were used in the field experiment, all located in the Vosges Mountains, North-eastern France, a region which has received high atmospheric acid deposition in the past (Party et al., 1995; Probst et al., 1999). Soils in the region vary between acid brown and podzolic, and are underlain by quartz enriched (thus weathering-resistant) granite or sandstone bedrock (Party et al., 1995). Due to small-scale differences in the mineral composition of the bedrock, nearby streams may have quite different pH, ANC, total Al and base cations concentrations (Dangles et al., 2004). Four nearby streams were selected along an acidification gradient on both granite and sandstone bedrock (Table 1) to evaluate how the magnitude of acidification effect on litter decomposition may depend on parent lithology and acidifica-

Table 1
Location and water characteristics (mean ± SD, n = 10) of the study streams during the litter decomposition experiment (December 18, 2008–February 26, 2009). Within each parent lithology (granite or sandstone), streams are ordered by increasing acidity with the first stream being a reference (circumneutral). ANC, acid neutralizing capacity.

Parent lithology and stream name	Stream acronym	Latitude (N)	Longitude (E)	Elevation (m asl)	Conductivity ($\mu S\ cm^{-1}$)	pH	ANC ($\mu eq\ L^{-1}$)	Total Al ($\mu g\ L^{-1}$)	Ca^{2+} ($mg\ L^{-1}$)	NO_3^- ($mg\ L^{-1}$)
Granite bedrock										
Tihay	TH	47°58′50.9″	6°52′32.6″	667	46.4 ± 4.7	6.65 ± 0.10	102 ± 23	53 ± 41	2.29 ± 0.34	2.83 ± 0.36
Grand-Clos	GC	47°58′46.3″	6°52′33.4″	647	16.9 ± 0.8	5.99 ± 0.09	21 ± 4	88 ± 27	0.85 ± 0.08	0.98 ± 0.16
Longfoigneux	LF	47°57′57.5″	6°51′53.3″	620	15.7 ± 0.6	5.49 ± 0.17	5 ± 3	128 ± 23	0.67 ± 0.08	0.78 ± 0.12
Wassongoutte	WA	47°58′27.0″	6°53′12.8″	668	14.2 ± 0.8	5.11 ± 0.17	−3 ± 3	188 ± 160	0.42 ± 0.08	0.84 ± 0.14
Sandstone bedrock										
La Maix	LM	48°27′58.9″	7°03′17.3″	387	80.5 ± 5.9	7.33 ± 0.09	523 ± 51	61 ± 77	7.05 ± 0.59	3.29 ± 0.13
Ravines	RV	48°25′14.8″	6°56′39.3″	382	35.4 ± 0.9	5.21 ± 0.11	0 ± 2	107 ± 42	1.75 ± 0.09	2.88 ± 0.11
Gentil Sapin	GS	48°27′03.7″	7°03′58.3″	536	30.6 ± 2.0	4.57 ± 0.14	−22 ± 10	413 ± 175	1.16 ± 0.19	4.45 ± 0.66
Basse des Escaliers	BE	48°27′58.9″	7°05′46.2″	740	30.8 ± 1.9	4.39 ± 0.09	−35 ± 7	571 ± 107	0.76 ± 0.05	2.67 ± 0.40

tion level. The streams have similar hydrology and morphology and channel substrate is composed of cobble, gravel, and coarse and fine sand. Mixed conifer-broadleaf forests dominated by silver fir *Abies alba* Mill., Norway spruce *Picea abies* L., and common beech *Fagus sylvatica* L. cover the area, with riparian zones being largely dominated by *F. sylvatica* and black alder *Alnus glutinosa* (L.) Gaertn. No anthropogenic activities exist upstream of the study sites in these small streams, besides low level forestry.

During the litter decomposition experiment, water chemistry was determined on each date when leaf bags were introduced into or retrieved from the streams. Water samples were collected from each stream in polyethylene bottles, transported to the laboratory in a cooler, and analyzed within 48 h. Water electrical conductivity was determined at 25 °C with a Metrohm E518 conductometer (Herisau, Switzerland), pH was determined with a microprocessor pH meter (pH 3000, WTW, Weilheim, Germany), and ANC was determined by Gran titration (Gran, 1952). Total Al (after acidification with nitric acid) and calcium (Ca^{2+}) concentrations were determined by atomic absorption spectrophotometry (Aanalyst 100, Perkin Elmer and Varian Spectr AA-300, Waltham, MA, USA) and nitrate (NO_3^-) concentration was determined by ion chromatography (Dionex 1500i with an AS 4 A SC column; Sunnyvale, CA, USA). Note that during the experiment, stream water pH showed little variation in circumneutral streams (Table 1) indicating that these streams (LM and TH) did not exhibit acid stress and can be considered as reference streams.

2.1.2. Leaf litter decomposition

In Autumn 2008, leaves of *A. glutinosa*, sycamore *Acer pseudoplatanus* L., and *F. sylvatica* were collected at abscission from the riparian vegetation of La Maix stream, air dried at room temperature and stored in the dark until needed. These species are present in the riparian zones of the study streams and were selected to represent a gradient of intrinsic litter characteristics and associated processing rates (Webster et al., 1995). Air dry leaves were weighed (3.00 ± 0.03 g, mean ± SE), moistened with distilled water to render them soft and less susceptible to breakage during handling and transport, and enclosed in fine mesh (0.5 mm mesh opening; prevents macroinvertebrate access to the litter) and coarse mesh (5 mm mesh opening; allows macroinvertebrate access to the litter) bags (10 × 15 cm). Note that before weighting, the petiole of *A. pseudoplatanus* was removed.

Litter bags (16 per species and mesh size) were deployed in each of the eight streams on December 18, 2008. Sampling (4 replicates per treatment) occurred on four occasions after 2–49 (*A. glutinosa*), 2–63 (*A. pseudoplatanus*), and 2–70 days incubation (*F. sylvatica*). Sampling dates and maximum incubation duration were selected a priori for each species taking into account expected decomposition rates so that at least 50% mass loss would be achieved by the last sampling date. After retrieval, litter bags were enclosed individually in plastic bags and transported to the laboratory in a cooler. In the laboratory, litter bags were opened above a 0.5-mm sieve to retain small litter fragments, and leaves rinsed with water from the corresponding stream. Litter mass remaining was oven dried at 105 °C until constant mass and weighed (± 0.01 mg) for the determination of dry mass (DM). Dry mass was ignited in a muffle furnace at 550 °C for 4 h and the ashes were weighed (± 0.01 mg). Ash free dry mass (AFDM) remaining on each sampling date was determined by the difference between DM and ash mass. Results were expressed as proportion of AFDM remaining, given by the ratio between final and initial litter AFDM. Initial (day 0) litter AFDM was estimated by applying a conversion factor between initial air dry mass and initial AFDM calculated from extra sets of leaf litter bags (n = 4 per leaf species and mesh size) that were taken to the field on day 0, returned to the laboratory, and processed as described above.

Decomposition rates (k, d^{-1}) were estimated by fitting negative exponential models to the proportion of litter AFDM remaining over time. The effect size of acidification on the decomposition rate was calculated as the response ratio R, which is given by the ratio of litter decomposition rate in the acidified stream (k_{acid}) to the litter decomposition rate in the reference stream (k_{ref}), $R = k_{acid}/k_{ref}$; R = 1 indicates no effect of acidification, R < 1 indicates inhibition and R > 1 indicates stimulation of litter decomposition in acidified streams (Ferreira et al., 2015). Analyses are performed on the natural logarithm of R (lnR) (Hedges et al., 1999; see Section 2.3.1)

2.2. Meta-analysis

2.2.1. Primary studies and case studies

We searched for primary studies that addressed the effect of acidification due to atmospheric acid deposition on litter decomposition in streams and rivers, and were published in scientific journals, in English, between January 1970 and April 2016. The search was done using Google Scholar, personal literature databases, and reference lists in primary studies. Search in Google Scholar was done using combinations of the following search terms: (decomposition or processing or breakdown or decay) and (litter or leaf or leaves or bark or wood) and (pH or acid or low alkalinity or low acid neutralizing capacity, ANC) and (stream or river or water course).

Primary studies ('studies' from here onwards) that satisfied the following criteria were included in the analysis: (i) address the effects of acidification due to atmospheric acid deposition on litter decomposition, (ii) focus on natural running waters (i.e., streams or rivers) rather than experimental stream channels or other manipulative approaches, (iii) compare litter decomposition for at least one circumneutral (reference) stream and one equivalent acidified stream, (iv) report decomposition of litter of allochthonous origin (i.e., tree leaves or woody substrates) that was incubated at the surface of the sediment (not in the hyporheic zone), (v) report sample size (n) and a measure of variation [standard error (SE), standard deviation (SD), 95% confidence limit (CL); not necessarily mandatory (see Section 2.2.2)] for both reference and acidified streams. Twenty studies satisfied these inclusion criteria, but three studies (Griffith and Perry, 1993; Dangles and Guérold, 2000; Clivot et al., 2014) were not included in the database due to multiple publication (i.e., the same data had been shown in two studies and thus one of these was excluded from the analysis to avoid data duplication; Appendix S1). Thus, the database was composed of 17 studies, which contributed 67 unique 'cases studies' (Table S1).

Each 'case study' ('case' from here onwards) consisted of a comparison between a reference stream and an acidified stream. When multiple reference streams and multiple acidified streams were reported in a study, reference–acidified pairs were specified after personal communication with the author (e.g., Clivot et al., 2013). When pairing reference and acidified streams was not possible due to lack of information or uneven number of streams of each type, reference streams were averaged and contrasted with each acidified stream (e.g., Dangles et al., 2004). When studies considered a gradient of acidification, the stream described as the reference or the least impacted (defined not only by the circumneutral pH, but possibly also by low Al concentration and high ANC) was contrasted with each acidified stream (e.g., Cornut et al., 2012). Also, a few studies compared different litter species (e.g., Griffith et al., 1995) or different mesh sizes (e.g., Mackay and Kersey, 1985) between reference and acidified streams. Thus, many studies contributed with several cases to the database (Table S1). Despite the lack of independency among several cases originating from the same study, we have included them in this analysis to enable assessment of associated moderators. We have also assessed the influence of including multiple cases per study via sensitivity analysis (see Section 2.3.2).

2.2.2. Effect size

The effect size of acidification on litter decomposition rate was calculated as the response ratio R, as described in Section 2.1.2; analyses are performed on the natural logarithm of R (lnR) (Hedges et al., 1999; see Section 2.3.2). In most cases, litter decomposition was reported as the decomposition rate (k, d^{-1}; negative exponential model), which was used directly in the calculation of the effect size. When litter decomposition was expressed as % litter mass remaining or % litter mass loss after a given incubation period, this was converted into decomposition rate (k, d^{-1}) assuming a negative exponential decay (e.g., Mackay and Kersey, 1985).

The variance associated with lnR (V_{lnR}), needed to weigh the effect sizes in the meta-analysis (see Section 2.3.2), was calculated from the SD and sample size (n) associated with each decomposition rate value (Borenstein et al., 2009). If variance was reported as SE or 95% CL, it was converted into SD. In the few cases where no measure of variance associated with decomposition rates was given in the study or provided by the authors, SD values were estimated by imputation based on the cases in the database that reported SD values associated with decomposition rates (Koricheva et al., 2013).

2.2.3. Moderator variables

Information on leaf litter identity (several genera), mesh size [coarse (> 0.5 mm mesh opening) and fine (≤0.5 mm mesh opening)], pH (used to estimate H^+ concentration) and Al concentration (both continuous variables) was retrieved from primary studies or requested from the authors (Table S1). These explanatory variables, referred to as moderators in meta-analysis, may affect the magnitude of the response of litter decomposition rate to acidification, and thus contribute to understanding the conditions in which litter decomposition may work best as a bioassessment tool of stream functional integrity in the face of acidification. Moderator variables will be used in subgroup analyses (categorical moderators) or meta-regressions (continuous moderators) (see Section 2.3.2).

2.3. Statistical analyses

2.3.1. Field study

Decomposition rates per leaf litter species and mesh size (considering all streams together) were related with water pH and total Al concentration (mean values during each litter incubation period) by exponential and power models, respectively.

The overall effect of acidification on litter decomposition and the effects of leaf litter species, mesh size and parent lithology on the magnitude and direction of litter decomposition response to acidification were assessed using meta-analytic techniques as described below (see Section 2.3.2).

2.3.2. Meta-analysis

A random-effects model of meta-analysis was used to determine the grand mean effect size, i.e., the overall effect of acidification on leaf litter decomposition; between-study variance was estimated by the restricted maximum likelihood (REML) method. Individual effect sizes (lnR) were weighted by the reciprocal of their variance to account for differences in accuracy among cases. To facilitate interpretation of the results, the mean effect size lnR was back-transformed into mean effect size R; a significant effect existed if the 95% CL did not include 1. The percentage of total variability that is due to between-study variation rather than sampling error (I^2) was also calculated (Borenstein et al., 2009).

The effects of moderators on the magnitude and direction of leaf litter decomposition response to acidification were assessed for subsets of the database according to our questions and available sample size. Mixed-effects models were used to compare heterogeneity between (Q_B) and within moderator levels to assess the significance of each categorical moderator, i.e., leaf litter identity and mesh size (Koricheva et al., 2013); only levels with at least three effect sizes were compared (Table S1). Two moderator levels were significantly different if their 95% CL did not overlap. Significant effects of levels within moderators existed if the 95% CL did not include 1. Meta-regressions were used to assess the relationship between effect sizes (lnR) and continuous moderators: magnitude (ln-transformed) of pH decrease (as increase in H^+ concentration) between reference and acidified streams [$\ln([H^+]_{acid}:[H^+]_{ref})$] and magnitude (ln-transformed) of Al increase in concentration between reference and acidified streams [$\ln([Al]_{acid}:[Al]_{ref})$].

Considering multiple cases per study may have biased our results and thus a sensitivity analysis was performed. A mean effect size per study was calculated as the weighed mean effect size of all cases considered within that study using mixed-effects model, and analyses were repeated to the extent possible considering a mean effect size per study.

Evidence of publication bias in the overall database and in the database considering a single effect size per study was assessed by the funnel plot, which plots effect sizes against precision (symmetrical distribution of effect sizes around the grand mean effect size indicates no publication bias). Evidence of publication bias in the overall database and subsets used for analyses was assessed by the Rosenberg's fail-safe number (Nfs), which gives the number of missing effect sizes showing no significant effect that would be needed to nullify the grand mean effect size. If Nfs $> 5 \times n + 10$, with n = number of effect sizes, the results can be considered robust despite the possibility of publication bias (Koricheva et al., 2013).

Statistical analyses were performed in RStudio (RStudio, 2012) with the metafor package (Viechtbauer, 2010).

3. Results

3.1. Field study

3.1.1. Leaf litter decomposition

The three leaf litter species lost mass over time and after 49 days incubation *A. glutinosa* litter had 36–71% (fine mesh) and 4–67% (coarse mesh) mass remaining across streams, after 63 days incubation *A. pseudoplatanus* litter had 41–74% (fine mesh) and 18–72% (coarse mesh) mass remaining and after 70 days incubation *F. sylvatica* litter had 76–88% (fine mesh) and 57–86% (coarse mesh) mass remaining. The mass remaining data fitted well the negative exponential model and exponential decomposition rates varied between 0.0060–0.0208 d^{-1} (fine mesh) and 0.0075–0.0688 d^{-1} (coarse mesh) for *A. glutinosa* litter across streams, between 0.0043–0.0139 d^{-1} (fine mesh) and 0.0052–0.0268 d^{-1} (coarse mesh) for *A. pseudoplatanus* litter and between 0.0017–0.0038 d^{-1} (fine mesh) and 0.0021–0.0088 d^{-1} (coarse mesh) for *F. sylvatica* litter (Fig. 1). The large variation in decomposition rates within each mesh size and litter species reflected the varying degrees by which litter decomposition was inhibited over the acidification gradient.

3.1.2. Leaf litter decomposition response to acidification

Across all streams, decomposition rates of all three leaf litter species in both mesh sizes decreased with increasing acidity (i.e., declining pH; exponential function, R^2: 0.83–0.96, $p < 0.001$) and total Al concentration (power function, R^2: 0.65–0.89, $p < 0.001$) (Fig. 2).

The grand mean effect size R was 0.36 (95% CL: 0.31–0.42; Fig. 3), which corresponds to a significant inhibition of leaf litter decomposition rates in acidified streams by an average of 64% (95% CL: 58% − 69%). Acidification significantly inhibited the decomposition of all three leaf litter species, in both mesh sizes and parent lithologies, by 52–73% (R = 0.27–0.48; Fig. 3). Litter decomposition was similarly inhibited for all three species (−68% overall for *A. glutinosa*, −65% for *A. pseudoplatanus* and −57% for *F. sylvatica*; $Q_B = 2.654$, df = 2, $p = 0.265$) (Fig. 3). The inhibition was significantly stronger in coarse (−73% overall) than in fine mesh bags (−52% overall) ($Q_B = 24.525$, df = 1, $p < 0.001$) and stronger in sandstone (−69% overall) than in granite bedrock (−58% overall) ($Q_B = 4.166$, df = 1, $p = 0.041$) (Fig. 3).

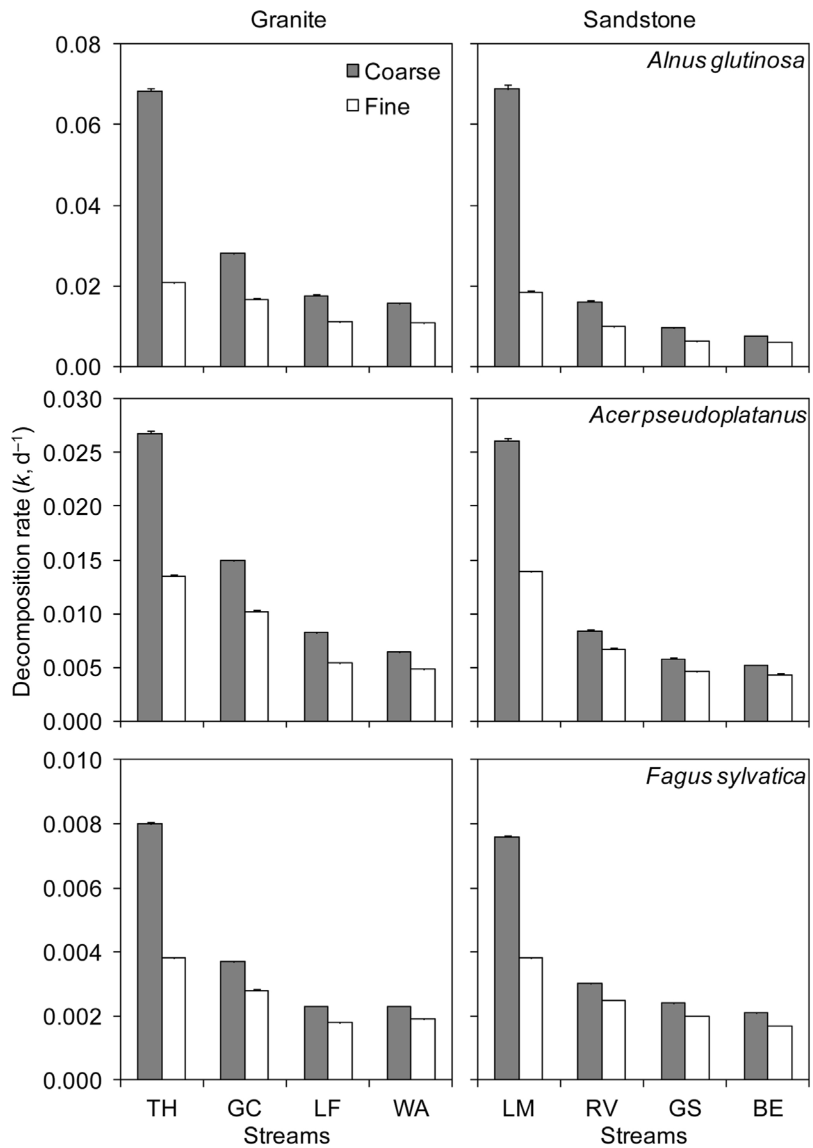

Fig. 1. Exponential decomposition rates of three leaf litter species enclosed in coarse and fine mesh bags and incubated in streams flowing through either granite or sandstone bedrock. Within each parent lithology (granite or sandstone), streams are ordered by increasing acidity with the first stream being a reference (circumneutral); for stream acronyms see Table 1. R^2: 0.75–0.99 and $p < 0.0001$ in all cases.

3.2. Meta-analysis

3.2.1. Database

The overall database consisted of 67 cases derived from 17 studies, being the earliest studies from 1985 (Kimmel et al., 1985; Mackay and Kersey, 1985) (Appendix S1, Table S1). All studies used leaf litter, mostly from *F. sylvatica* (45% of cases) and *Acer* sp. (30% of cases), which was generally incubated in coarse mesh bags (79% of cases) (Table S1).

3.2.2. Overall leaf litter decomposition response to acidification

The grand mean effect size R was 0.37 (95% CL: 0.30–0.46; Fig. 4), which corresponds to a significant inhibition of leaf litter decomposition rates in acidified streams by an average of 63% (95% CL: 54%–70%). Assessment of the grand mean effect size was not strongly influenced by the lack of independency as its value did not change much when each study was represented by a single effect size (R = 0.37, 95% CL: 0.26–0.51). The overall database (n = 67 cases) and the database considering a single effect size per study (n = 17 studies), did not seem to be affected by publication bias as the funnel plot was symmetrical in both cases. The Nfs were also above the threshold for considering the results robust in both cases (overall database, Nfs = 312 700, Nfs > 345; single study database, Nfs = 175, Nfs > 95). The percentage of total variation in the overall database that was explained by between-study variation was high (I^2 = 99%), suggesting that differences in methodological approaches (e.g., mesh size or leaf litter species used) and environmental conditions likely moderate the response of leaf litter decomposition to acidification.

3.2.3. Effects of moderator variables

The response of litter decomposition to acidification depended on leaf litter identity (Q_B = 28.808, df = 4, $p < 0.0001$; Fig. 4). The inhibition of decomposition rates in acidified streams was significant for *Fagus* (−78%), *Alnus* (−58%) and *Acer* (−47%), but not for *Quercus* and *Liriodendron* (but sample size was small). Mean effect size for litter species declined directionally among all leaf types, but comparatively large variation around most species rendered the effect size associated with the greatest degree of inhibition (i.e., *Fagus*, R = 0.22) as distinct from *Acer*, *Quercus* and *Liriodendron*, the three species with the smallest inhibition. The inhibition of litter decomposition in acidified streams was significant for both mesh sizes, but the magnitude of the response

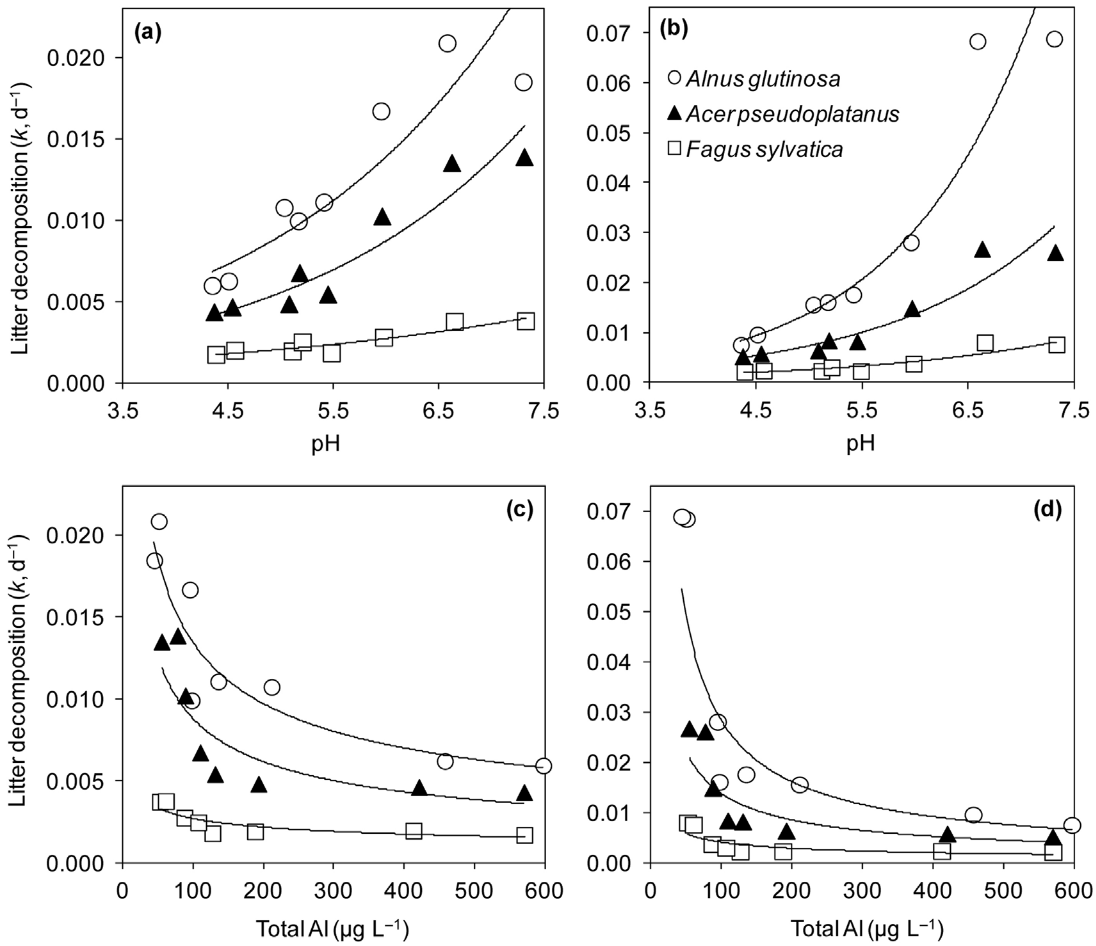

Fig. 2. Relationship between the decomposition rates of three leaf litter species enclosed in fine mesh (a, c) and coarse mesh (b, d) bags and pH (a, b) and total Al concentration (c, d). Streams flowing through granite or sandstone bedrock were considered together. The fit of the positive exponential models (pH; R^2: 0.83–0.96, $p < 0.001$) and negative power models (total Al; R^2: 0.65–0.89, $p < 0.001$) is also shown.

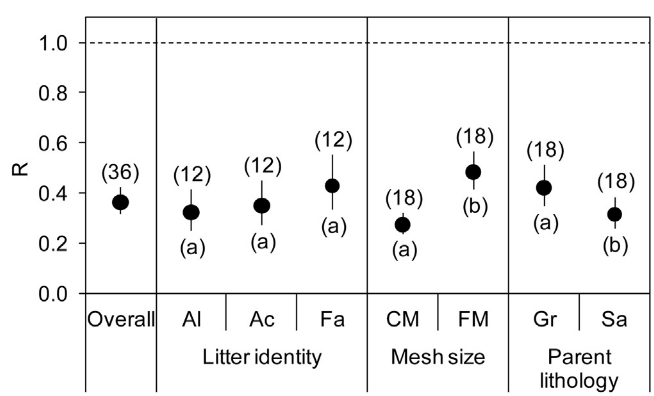

Fig. 3. Effect of acidification (R ± 95% CL) on leaf litter decomposition in the field study, overall and as a function of leaf litter identity, mesh size and parent lithology. The dashed line (mean effect size R = 1) indicates no effect of acidification on litter decomposition and mean effect size R < 1 indicates inhibition of litter decomposition in acidified streams. The effect of acidification is significant when the 95% CL does not include 1. For each moderator, levels with the same letter do not significantly differ in their response to acidification. Values in parenthesis indicate the sample size. Al, *Alnus glutinosa*; Ac, *Acer pseudoplatanus*; Fa, *Fagus sylvatica*; CM, Coarse mesh; FM, Fine mesh; Gr, Granite; Sa, Sandstone.

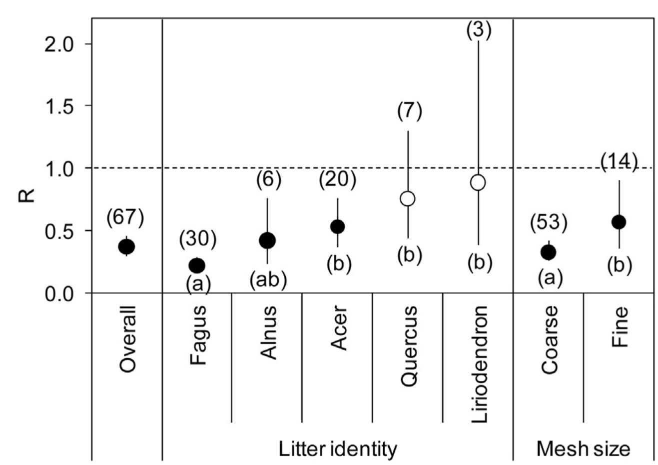

Fig. 4. Effect of acidification (R ± 95% CL) on leaf litter decomposition in the meta-analysis, overall and as a function of leaf litter identity and mesh size. The dashed line (mean effect size R = 1) indicates no effect of acidification on litter decomposition, mean effect size R > 1 indicates stimulation and mean effect size R < 1 indicates inhibition of litter decomposition in acidified streams. The effect of acidification is significant when the 95% CL does not include 1 (black circles). For each moderator, levels with the same letter do not significantly differ in their response to acidification. Values in parenthesis indicate the sample size. Nfs for each dataset used were as follow: litter identity, Nfs = 308 044 (n = 66, one case using *Fraxinus* was not considered); mesh size, Nfs = 312 700 (n = 67, the entire database was used).

was significantly stronger for coarse mesh (−67%) than for fine mesh (–43%) bags ($Q_B = 4.251$, df = 1, $p = 0.039$) (Fig. 4).

The response of leaf litter decomposition to acidification did not depend on the magnitude of the increase in H^+ concentration between reference and acidified streams ($p = 0.936$; Fig. 5a). On the other hand,

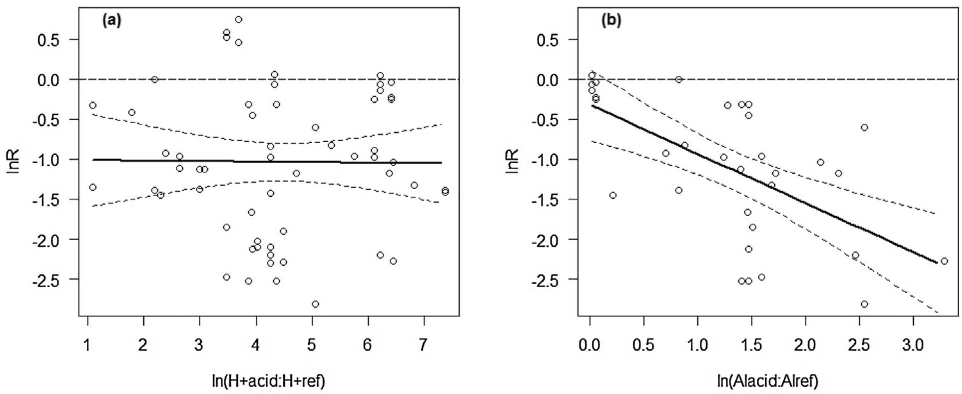

Fig. 5. Effect of the magnitude of the increase in H^+ (a) and in Al concentration (b) between reference and acidified streams on leaf litter decomposition (lnR). The dashed line (lnR = 0) indicates no effect of acidification on litter decomposition, lnR > 0 indicates a stimulation and lnR < 0 indicates an inhibition of litter decomposition in acidified streams. The relationship (meta-regression) is shown by the solid line and the 95% CL by the dashed lines.

effect sizes decreased substantially and significantly with increasing Al enrichment in acidified vs. reference streams (slope = −0.614, $p < 0.0001$; Fig. 5b).

4. Discussion

Although signs of chemical recovery from acidification have been reported in many areas throughout Europe and North-America, biological recovery remains poor and chronic or episodic acidification still remains an ecological threat (Dunford et al., 2012; Pye et al., 2012). Thus, it is crucial to assess the degree to which streams functional integrity in acid-sensitive regions is impaired as this influences their ability to provide ecosystem services. Leaf litter decomposition has been proposed as a bioassessment tool for stream functional integrity in the face of human influences (Gessner and Chauvet, 2002; Young et al., 2008), including acidification (Dangles et al., 2004; Simon et al., 2009). Indeed, both our field study and meta-analysis revealed strong inhibition of leaf litter decomposition in acidified streams. The magnitude of this response depended on methodological characteristics (mostly mesh size) and water chemistry, as anticipated.

4.1. Leaf litter decomposition was strongly inhibited in acidified streams

The results from the field study were consistent with those from the meta-analysis, and both revealed strong overall inhibition (> 60%) of leaf litter decomposition across acidified streams. This response was expected based on previous extensive reports of altered microbial and macroinvertebrate community structure and activity, and reduced litter decomposition in acidified streams (e.g., Dangles and Guérold, 2001a,b; Dangles et al., 2004; Simon et al., 2009; Cornut et al., 2012; Clivot et al., 2013). The reported responses of community structure and activity and of litter decomposition to concomitant decreases in pH and base cations concentrations, and increases in Al concentration in acidified streams are robustly consistent across studies and may reflect an 'acidification syndrome'. The biological symptoms of this syndrome are manifested in microbial and animal composition, abundance, and activity. In the microbial realm, this includes lower aquatic hyphomycete species richness (Baudoin et al., 2008; Cornut et al., 2012), microbial respiration (Dangles et al., 2004; Simon et al., 2009), and fungal biomass (Griffith and Perry, 1994; Dangles et al., 2004; but see Dangles and Chauvet, 2003; Baudoin et al., 2008; Clivot et al., 2013), along with altered microbial enzymatic activities (Griffith et al., 1995; Clivot et al., 2013). For macroinvertebrate consumers, the syndrome entails reductions in abundance or even disappearance of acid-sensitive macroinvertebrate species, including gammarid, sericostomatid and limnephilid detritivores, and their replacement by smaller and less efficient shredders (e.g., leuctrids) (Kimmel et al., 1985; Meegan et al., 1996; Dangles and Guérold, 1998, 2001a,b; Guérold et al., 2000; Tixier and Guérold, 2005; Simon et al., 2009), and reductions in shredder biomass (Meegan et al., 1996; Dangles and Guérold, 1998). All of which result in the well documented reduction in litter decomposition rates (present meta-analysis).

4.2. The inhibition of leaf litter decomposition was not strongly affected by litter identity

Contrary to our prediction, however, the response of litter decomposition to acidification did not differ among leaf litter species in the field study, despite differences in litter characteristics among species (Webster et al., 1995). In the meta-analysis, the inhibition of litter decomposition in acidified streams was stronger for *Fagus* than for *Acer*, *Quercus* and *Liriondendron*, but it did not differ when compared with *Alnus*. The field study and meta-analysis appear to present conflicting results. We are, however, more confident in 'study-derived evidence' than in 'review-derived evidence' since, on one hand, our field study was the study most extensively comparing the response of different litter species to acidification (i.e., three litter species in two mesh sizes along an acidification gradient over two parent lithologies; see also Griffith and Perry, 1994; Griffith et al., 1995; Meegan et al., 1996) and, on the other hand, differences among litter species revealed in the meta-analysis may be confounded by differences in environmental characteristics among studies leading to the high degree of variability observed for a given litter species. Overall, however, the absence of strong differences in the response of leaf litter decomposition to acidification among litter species suggests that decomposers are likely inhibited to similar degree on all substrates.

4.3. The inhibition of leaf litter decomposition was stronger in coarse mesh than in fine mesh bags

In agreement with our prediction, the influence of acidification on litter decomposition was much stronger in coarse mesh than in fine mesh bags, both for the field study and meta-analysis. This is congruent with the fact that acidification strongly reduces the abundance and biomass of acid-sensitive macroinvertebrate species, including gammarids, sericostomatids and limnephilids that are large and efficient shredders (Meegan et al., 1996; Dangles and Guérold, 1998, 2001a,b; Simon et al., 2009). Thus, macroinvertebrate-driven litter mass loss in acidified streams is reduced both indirectly via a decrease in microbial activity (reflecting shredders preference for fully conditioned leaf litter; Bärlocher and Sridhar, 2014) and directly via a decrease in shredder efficiency and biomass, which results in stronger decreases in litter decomposition in coarse mesh than in fine mesh bags (Mackay and Kersey, 1985; Dangles and Guérold, 2001b).

4.4. The inhibition of leaf litter decomposition was robust to differences in parent lithology

The inhibition of litter decomposition in acidified streams was stronger in sandstone than in granite bedrock in the field study, likely due to the larger acidification gradient and the larger differences in water pH and Al concentrations between the reference stream and the acidified streams in the former than in the later bedrock type. This was supported by the observation that litter decomposition rates were closely relate to pH across streams where the collection of sites generated a robust acid gradient. The direction of the response of leaf litter decomposition to acidification was thus consistent independent of parent lithology, confirming that inhibition of litter decomposition is a general symptom of the acidification syndrome.

4.5. The inhibition of leaf litter decomposition was stronger where the degree of acidification was higher

The magnitude of the response of leaf litter decomposition to acidification increased with increasing degree of acidification, i.e., decrease in pH (or increase in H^+ concentration; field study) and increase in Al concentration (field study and meta-analysis), as expected based on previous findings (Dangles et al., 2004; Simon et al., 2009; Cornut et al., 2012). Increases in the level of acidification, which imply decreases in pH and in base cations concentrations and increases in Al concentration (chemical symptoms of the acidification syndrome), promote stronger inhibition of microbial and invertebrate activities and thus stronger reduction in litter mass loss (Dangles et al., 2004; Cornut et al., 2012).

4.6. Leaf litter decomposition as a bioassessment tool of acidification effects in streams

The field study showed that leaf litter decomposition was strongly inhibited in acidified streams and that this response was consistent across leaf species, mesh sizes and parent lithologies, and the meta-analysis showed that these results are robust and consistent across the wider range of conditions tested in previous studies. The magnitude of inhibition increased with the acidification degree in the field study, and the meta-analysis showed that this is a general trend. Together, these results support the suggestion that leaf litter decomposition has strong potential to be used as an assessment tool for acidification effects on stream ecosystem functioning.

The use of litter decomposition as an assessment tool for acidification is supported by sound theoretical ecological concepts since litter decomposition is a fundamental ecosystem process that integrates the activities of an array of organisms and changes in environmental conditions (e.g., water chemistry). The response of leaf litter decomposition to acidification allows for a priori predictions since litter decomposition is consistently inhibited in acidified streams and the magnitude of this inhibition can be used to indicate the degree of impairment, as shown in this study. The implementation of litter decomposition protocols is relatively simple and cheap when compared with protocols for sampling, sorting and identifying structural elements, and can be easily standardized and applied over large spatial scales (Gessner and Chauvet, 2002; Bonada et al., 2006). In fact, the meta-analysis showed precisely this as the effect of acidification on leaf litter decomposition was consistent across all the 17 studies considered.

The inhibition of leaf litter decomposition was, nevertheless, stronger for coarse mesh than for fine mesh bags, but since the response was already so strong for fine mesh bags, any mesh size can be used for assessing acidification effects on leaf litter decomposition. However, if the potential for physical abrasion is great (e.g., high current velocity or sediment in transport) and litter mass loss due to physical fragmentation is a strong possibility, coarse mesh bags should not be used. On the other hand, coarse mesh bags should be used if effects mediated by changes in the macroinvertebrate community are targeted.

Gessner and Chauvet (2002) proposed the ratio between the litter decomposition rate in an impacted site to the litter decomposition rate in the reference site (k_{imp}/k_{ref}) as one possible metric to assess stream functional integrity, with three classes indicating (i) no clear evidence of impact when $k_{imp}/k_{ref} = 0.75–1.33$, (ii) some evidence of compromised ecosystem functioning when $k_{imp}/k_{ref} = 0.50–0.75$ or 1.33–2.0, and (iii) severely compromised ecosystem functioning when $k_{imp}/k_{ref} < 0.5$ or > 2. This metric is, in fact, an effect size and matches exactly the one employed here ($R = k_{acid}/k_{ref}$). However, if applying the classes tentatively proposed by Gessner and Chauvet (2002), Grand-Clos stream (GC) in the field study would be classified as having no clear evidence of impact when considering *A. glutinosa* and *A. pseudoplatanus* litter decomposition in fine mesh bags (R = 0.80 and 0.76, respectively). However, when using the effect size approach and its corresponding 95% CL, we find that litter decomposition in GC is significantly inhibited in relation to the reference stream and thus there is evidence of compromised ecosystem functioning. We would, thus, propose that the effect size concept (together with its corresponding 95% CL) be used to identify acidification effects on litter decomposition.

5. Conclusion

The inhibition of leaf litter decomposition is clearly tied to an 'acidification syndrome' that promotes well-known and repeated changes such that litter decomposition can be a powerful tool to detect acidification effects on stream ecosystem functioning. As well, leaf litter decomposition will likely respond to improvements in water chemistry as those expected from the ongoing decrease in atmospheric deposition of acidifying substances, or from bioremediation programs (e.g., liming) (Merrix et al., 2006). Thus, it represents an appropriate tool to assess biological recovery as well as to determine ecological status according to the European Water Framework Directive (2000/60/EC; European Parliament and Council, 2000). Since the ability of an ecosystem to provide services depends on intact functioning, monitoring programs should consider the inclusion of functional tools proven to be consistent in response to impacts and across regions, and robustly independent of experimental choices (e.g., mesh size or litter species).

Acknowledgements

We thank P. Gierlinski, P. Wagner and P. Rousselle for field and laboratory assistance. We also thank authors for providing information that was not available in the primary studies used in the meta-analysis and two anonymous reviewers for their helpful comments on an earlier version of the manuscript. The present study was financed by a French ANR program (ANR-07-BDIV-007-01 Recover) and the Zone Atelier Moselle (LTER France). Support given by the Portuguese Foundation for Science and Technology (FCT), through the strategic project UID/MAR/04292/2013 granted to MARE and through financial support given to VF (IF/00129/2014) is gratefully acknowledged.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ecolind.2017.04.044.

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