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A Cross-System Analysis of Litter Chemical Dynamics Throughout Decomposition

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Abstract

Decomposition of plant litter is a fundamental ecological process, integral to soil formation, soil organic matter chemistry, and biogeochemical cycling. However, much of our understanding of decay dynamics focuses on rates of litter mass loss and therefore carbon dynamics, with relatively less exploration of the chemical nature of litter decomposition during which the degradation of litter structural and metabolic compounds into fragments are either metabolized or ultimately incorporated into soil humus. Our understanding of the patterns of changes in litter chemistry throughout decomposition is incomplete, as few studies have measured chemical content beyond initial litter chemistry and throughout decay, and particularly not chemistry beyond carbon and nitrogen. The existing literature also reports idiosyncratic instances of litter chemical convergence and divergence. We used archived litter decomposition samples and data from across the U.S. Long-Term Ecological Research Network to investigate the trajectory of a comprehensive array of litter chemistry, including nutrient, structural, and metabolic parameters, across a wide variety of plant functional types and ecosystems, throughout the first 70% of mass loss. Our results do not yield a universally common pattern of litter chemical trajectories across all functional types and regions, and very limited evidence of convergence or divergence in chemistry over time, mostly within the nutrient elements. We provide details about the behavior of individual chemical parameters to functional type and region over decay. Changes in plant communities driven by global change may alter nutrient cycling and SOM formation through persistence or divergence on litter chemistry inputs.

Key words: litter decomposition; litter chemistry; litter quality; plant functional types; cross-system synthesis; nutrients; py-GCMS.

Highlights

- No common pattern of litter chemistry exists across plant species or ecosystems
- We find no evidence of convergence in litter chemistry through 70% of decay
- Influence of functional type and environment differs across chemical parameters

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Author Contributions BB, LC, KW conceived and designed the study, and performed the sample analysis. BB led the data analysis and authorship of the paper, and LC and KW provided substantial input and feedback.

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Introduction

Over half of terrestrial net primary production (NPP) ends up in soil via decomposition (Wardle and others 2004). Decomposition of plant litter is thus a fundamental ecological process, integral to energy flow in food webs and nutrient cycling (Swift and others 1979). As such, decades of decomposition research have yielded rich information about the dynamics of litter mass loss over time, greatly informing our understanding of carbon (C) storage and mineralization in ecosystems. Relatively less is understood about the chemical dynamics of litter throughout decomposition, particularly rates of micronutrient element release and the degradation of litter structural and metabolic compounds whose fragments are either broken down during C and nutrient recycling or ultimately incorporated into SOM. Recent analytical capabilities now allow us to learn more about the chemical composition of these complex compounds, improving our understanding of how they change as they undergo decomposition in the stages leading up to SOM formation.

Extensive research on litter decomposition across ecosystems and plant species has provided ample consensus that litter chemistry acts as a main driver of decomposition rate across biomes, coupled with climate factors including temperature and moisture. For example, results from the TeaComposition initiative comparing decay of high chemical quality (high in nutrients and soluble compounds) green tea with low quality (nutrient-poor and high in recalcitrant compounds) rooibos tea shows that, after several months of decomposition over which 30–60% of litter mass was lost, 65% of variability in decomposition rate was related to litter type, with a positive correlation between mass loss and mean annual precipitation (Djukic and others 2018; Fanin and others 2019). The Nutrient Network (NutNet) also reported that litter quality, namely nutrient content and prevalence of soluble versus recalcitrant compounds, was a main driver for two sites, outranked only by biome effects of grassland versus forest (Ochoa-Hueso and others 2019). Similarly, initial litter chemistry and litter identity more strongly influenced mass and nitrogen (N) loss than climate or ecosystem type along two gradients in Europe (Hoeber and others 2020; Zhou and others 2020) and the Long-Term Intersite Decomposition Experiment (LIDET; Parton and others 2007; Li and others 2015). Even green leaf chemical and morphological traits can be excellent predictors of decay rate (Rosenfield and others 2020).

Interactions between litter chemistry and the decomposition environment are also repeatedly demonstrated in the literature. Studies suggest that decomposition of high-quality litter is more climate-sensitive (Fanin and others 2019), and decay rate in colder ecosystems is more climate-sensitive (Bélanger and others 2019). The trajectories followed by distinct litter chemical parameters, such as lipids and carbohydrates during decay can also differ by temperature (Kohl and others 2021). Ecosystem properties can also interact with litter quality to influence decay. For instance, soil nutrient availability is important for low-quality litter mass loss (Bonanomi and others 2017; Fanin and others 2019), and detritivorous invertebrates can alter litter decomposition beyond the effects of litter quality alone (Guo and others 2020).

Most studies have focused primarily on initial litter chemistry as the indicator for how litter will decay, and do not focus on the dynamics of the chemistry itself as the litter decays. Such studies have certainly improved our predictions of decomposition dynamics, including decay rate and nutrient release (for example, Cornwell and others 2008; Fyllas and others 2020). However, our current understanding of the patterns of changes in litter chemistry throughout decomposition is incomplete, partly because only a subset of studies has measured chemical content of litter throughout decay, and particularly not chemistry beyond C and N. Although measurements of litter chemistry during later stages of decay (beyond initial chemistry) have increased in recent years, the vast majority measure only %N, whereas %P is measured in less than half of them (for example, Killingbeck and others 1982; Schowalter and others 1998; Hernández and Hobbie 2008; Richardson and others 2010), and other elements much less frequently (for example, Killingbeck and others 1982; Zou and others 1995; Schowalter and others 1998; Lovett and others 2016). Numerous studies also measure lignin (for example, Moorhead and Reynolds 1993; Hunter and others 2003; Brandt and others 2010; Richardson and others 2010), but far fewer measure other structural and metabolic compounds (for example, Sullivan and others 1999; Filley and others 2008; Preston and others 2009; Wang and others 2021). Although initial chemistry may be a sufficient predictor for some chemical parameters (as is the case for N in Parton and others 2007), it is an untested hypothesis that initial chemistry determines release patterns for the majority of chemical parameters.

There are currently two general hypotheses that consider the nature of litter chemistry change

through time: chemical homogenization (convergence) versus development of distinct chemical identity (divergence). Evidence has informed hypotheses that litter chemistry becomes indistinguishable (convergence) after passing through the 'decay filter', representing the limited set of physiological capabilities of the decomposer community (Melillo and others 1989; Homann 2012; Wickings and others 2012). Alternatively, litter chemistry could diverge throughout decay, where initial differences become even greater as individual chemical parameters move along their individual trajectories to exacerbate initial differences. These alternative hypotheses are difficult to test because not many studies measure beyond initial chemistry, and when they do, do not typically cover a comprehensive array of chemical parameters. While some studies have investigated litter chemistry throughout decomposition at individual sites using litters from a limited range of initial chemistry (for example, Moorhead and Reynolds 1993; Ball and others 2009; Wallenstein and others 2013; Chen and others 2020), it is difficult to make comparisons across ecosystems due to the variety of chemical parameters measured and analytical methods used. The few studies examining changes in litter chemistry beyond C, N, and P have found little consistency in litter chemistry patterns during decomposition.

From the existing literature, it appears that the prevalence of litter chemical convergence and divergence depends on the chemical parameters under consideration. For example, compound classes determined by NMR and py-GCMS show some chemical traits converging and others diverging throughout decay (Wickings and others 2012; Wang and others 2019). Similarly, both divergence and convergence in litter nutrient content and stoichiometric ratios have been reported, with different patterns across studies (Moore and others 2011; Liu and others 2016; van Huysen and others 2016; Du and others 2020), and changes over time (Homann 2012). Further, studies show that the occurrence of convergence and divergence depends on environmental parameters and litter traits that vary across species. For example, the convergence in nutrient ratios observed by Liu and others (2016) was not true for all species, and there was an overall divergence in NMR-measured C biochemistry that depended on litter type, leading to no overall pattern in C biochemistry according to duration of decomposition. Using pyrolysis-based methods, Wickings and others (2012) and Wallenstein and others (2013) found that, even after extensive decay, litter chemistry was sensitive to differences in initial quality, land use intensity, and biological activity. Taken individually, these studies are understandably limited to just a few species and only a subset of litter chemistry. Studies that investigate compound classes rarely incorporate nutrients (particularly beyond N), and studies of nutrient dynamics rarely incorporate structural and metabolic compounds (particularly beyond lignin).

To date, it remains unclear whether the suite of litter chemical characteristics known to influence decomposition follow consistent patterns throughout decay across geographic space (and subsequently the climate, soil characteristics, and plant and decomposer communities native to that area). Better understanding of litter chemical dynamics throughout decomposition will aid our prediction of nutrient recycling and C dynamics in the litter layer, improving current models of ecosystem biogeochemistry. This better understanding is increasingly important in the context of global change, which is driving alterations in vegetation and decomposer communities (Kardol and others 2010; Nielsen and Ball 2015; Franklin and others 2016; Komatsu and others 2019). Altered presence and abundance of different plant species and functional types, with their characteristic leaf chemistry traits, will have implications for decomposition processes, and ultimately C cycling. For example, tree species and their associated litter chemical quality strongly influence N cycling and decomposition rates in forested ecosystems (Mudrick and others 1994; Finzi and Canham 1998; Lovett and Mitchell 2004), and their altered abundance will therefore influence litter decay dynamics. Understanding how various litter types might differ in their chemical parameters becomes important for predictions of future biogeochemistry.

The goal of this study was to investigate the trajectory of litter chemistry, including nutrient, structural, and metabolic parameters, using a comprehensive set of techniques, across a variety of species and ecosystems, throughout decomposition. To address this goal, we solicited archived litter decomposition samples from across the United States, largely within the Long-Term Ecological Research (LTER) Network, and analyzed a comprehensive and consistent set of litter chemistry. We combined this with compiled litter chemistry data from published studies, creating a dataset comprised of numerous litter species representing multiple plant functional types decomposed across many ecosystems. Plant functional types (PFTs) allow the categorization of species according to

similar characteristics to reduce the complexity of large-scale studies of numerous plant species (Duckworth and others 2000). Because PFTs differ in litter chemical and physical properties, they likely differ in decomposition dynamics (Silver and Miya 2001; Reich and others 2003; Cornwell and others 2008; Hoorens and others 2010). Choosing archived samples decomposed in different ecosystems also allowed us to evaluate the potential impacts of biotic and abiotic controls in interaction with litter chemical traits on decomposition. Using these data, we sought to identify whether there are universal patterns of changes in litter chemistry over decomposition, using a diverse but consistent set of chemical analyses. Specifically, we address the following questions: (1) Are there any consistent patterns in how litter chemistry changes throughout decay across geographic regions and plant functional types? In other words, does convergence or divergence of litter chemistry occur, and is it consistent across ecosystems and litter types that vary in initial litter chemistry? (2) Are these patterns consistent across litter chemistry parameters? In other words, if convergence and divergence are detected, does it happen across nutrient, structural, and metabolic chemistry, or do chemical categories differ in their pattern?

Methods

Collection of Archived Litter

To create a litter chemistry dataset that measures a consistent set of chemical parameters across a consistent set of decay stages, we solicited archived litter samples from prior litterbag studies of decomposition conducted in a variety of ecosystems that met the following criteria: (1) Only non-woody aboveground plant parts (leaves & stems) from single species decomposing alone, given that litter mixtures would introduce an extra level of complexity not able to be effectively addressed in this study; (2) Decomposed in an ecosystem where the litter was (or could reasonably have been) generated, such as species decomposed in their native ecosystem or agricultural species decomposed in an old field setting; (3) Decomposed on the soil surface in locations without synthetic inputs, such as fertilizer, irrigation, or chemical treatments.

We requested litter from only the retrieval dates that represented specifically targeted proportions of dry mass remaining: 100% (initial litter), 75% (early-stage decomposition), 50% (mid-stage of decomposition), and 30% (late stage of decomposition, given that few studies measure decomposition beyond this point). We recorded the actual proportion of mass remaining for each litterbag, but also assigned each to a ''bin'' representing these four target mass remaining stages (referred to as ''Stage Bin''). We considered the litter sample to be part of one of the bins as long as the average mass remaining of the replicates for that sampling date was within a proportion of 0.125 of the closest appropriate stage bin (in other words, if it was within the halfway mark to the next stage bin). The 0.125 range allowed for the fair inclusion of outliers, but we note that 75% of samples were within a proportion of 0.05. We used dry mass remaining to determine placement in these bins, given that all studies reported dry mass remaining, while only some measured ash-free dry mass (AFDM) remaining and did not always have extra material available to add these measurements. The duration of decomposition to achieve the target stages varied across the archived projects, and the days of decomposition were recorded for each sample along with the month (and therefore season) of each collection.

We analyzed up to four replicates from each project, as available. For studies where more than four replicates were available, we chose the four replicates that were closest to the targeted decay stage bin. Fewer than four replicates were included only if either the remaining archived mass was too low for chemical analyses in one of the replicates, or more commonly, the variability among the replicates caused some to not fall within the desired decay stage bin.

Collected samples were from studies that utilized a variety of mesh sizes. The actual mesh size was recorded, and was then categorized as ''small'' ($\leq$ 0.5 mm), ''medium'' (0.9–2 mm), or ''large'' (5–25 mm). Mesh sizes not included in these ranges were not represented in the archived samples. Litterbags were made from a variety of non-reactive materials, including Aclar plastic, carbon fiber, fiberglass, and nylon. We also categorized each study according to its climate group (tropical, subtropical, temperate, arid, Arctic), geographic region within the U.S. (northeast, southeast, midwest, southwest, and tropics), and the plant species and functional type. Plant functional group was assigned at a coarse scale (woody plants, herbaceous plants, or cactus) as well as at a fine scale where the coarse ''woody'' group was further differentiated into broadleaf (incorporating both deciduous and evergreen trees and shrubs) or coniferous; and the coarse ''herbaceous'' group into grass, sedge, or forbs. The cactus group was not further delineated.

For each project, we assigned a land use of the decomposition setting (native, urban, active agriculture, or old field agriculture).

Analysis of Archived Litter

Each collected litter sample was analyzed for carbon, macronutrients, micronutrients, and numerous classes of structural and metabolic compounds, unless it had been previously measured during the original study using a comparable method. If not previously ground prior to archival, litter samples were ground to a fine powder using a SPEC-Geno-Grinder®.

Total elemental % C and % N were determined on a Carlo-Erba NA1500® analyzer. Percent P, Ca, K, and Mg were measured using a dry ash acid digestion method in which 0.5 g of ground litter (or less if necessary due to low sample availability) was ashed in a muffle oven that was gradually brought to 475 °C over 1.5 h, held at 475 °C for 4 h, then dropped to 105 °C until digested in 5 ml of 35% HNO_3. Samples were then centrifuged at 25,250 × g for 10 min, and the supernatant diluted to 5% HNO_3 for measurement using inductively coupled plasma optical emissions spectroscopy (ICP-OES; Thermo iCAP6300, Hudson NH). Litter mass before and after the ashing procedure (prior to acid digestion) was used to calculate AFDM on samples for which AFDM remaining was not previously calculated in its original study.

Fiber compound categories were measured using sequential acid digestion (Van Soest, 1994). Briefly, 0.5 g of ground sample was placed into fiber filter bags and then digested in a fiber analyzer (ANKOM A200 Fiber Analyzer, Macedon, New York, USA) using first a neutral detergent to dissolve soluble (NDS) compounds, followed by low-normality H_2SO_4 detergent to dissolve acid-detergent soluble compounds (ADS, frequently correlated with hemicellulose), then 72% sulfuric acid to dissolve H_2SO_4 acid-soluble compounds (AS, frequently correlated with cellulose). The remaining material was identified as acid-indigestible (AI, frequently correlated with lignin). Filter bags were then ashed at 500 °C for 5.5 h to correct the fiber content for non-organic recalcitrant particles. Each compound category was then expressed as % of initial litter mass.

Samples were also analyzed for molecular composition using pyrolysis–gas chromatography and mass spectrometry (py-GCMS). Samples were first pyrolyzed on a CDS Pyroprobe 5150 pyrolyzer at 600 °C for 20 s (CDS Analytical, Inc., Oxford, PA, USA). Pyrolysis products were then transferred automatically to a Thermo Trace GC Ultra gas chromatograph (Thermo Fisher Scientific, Austin, TX, USA) and Polaris Q ion trap mass spectrometer (Thermo Fisher Scientific). Mass spectra were analyzed using Automated Mass Spectral Deconvolution and Identification System (AMDIS, V 2.65) and the National Institute of Standards and Technology (NIST) compound library. Compound abundances were calculated relative to the total ion signal from all detected and identified peaks. Individual compounds were analyzed separately and also arranged into the following functional groups: lignin, aromatic, phenols, polysaccharides, proteins, other nitrogen-bearing compounds, lipids, and compounds of unknown origin (Grandy and others 2009; Wickings and others 2011).

Data Hygiene

For the analyses reported here, we used only studies for which both 100% and 30% remaining (initial and late-stage) was available, most of which also included the intermediate 75% and 50% stages. Therefore, 75 and 50 were not equally represented in the replicates. In doing so, data were reduced to only those listed in Table 1. These criteria resulted in the collection of 39 different litter decay projects (defined as a particular species at a particular site in a particular experiment; Table 1). This includes three instances where the same species was decomposed in two separate projects at the same site (and thus listed only once in the table, rather than twice). The criteria eliminated the samples we collected from Arctic ecosystems/climates and sedges from the analyses. Across the included studies, the length of time to reach 30% mass remaining varied from 39 days (grass species decomposing in a midwest agricultural field) to 6 years (broadleaf species decomposing in the northeast). The vast majority of samples were from medium-sized mesh, providing an overall fairly consistent mesh size across the studies (Table 1).

Of the 39 projects, cactus was only decomposed in one region (the southwest). All other functional groups are present in at least two regions, with broadleaf woody plants as the only functional group to be represented across all regions. The tropics only had one functional group represented (broadleaf woody plants). All other regions contained at least two functional groups, and no region contained all functional groups. The following region-functional type combinations are represented by just one project (that is, only one species from one study): coniferous woody plants in the Midwest, and cactus, forbs, and broadleaf woody plants

Table 1. Plant Litter Decomposition Samples Used in Cross-site Analysis

Species	Region	Site	Functional Type	100	75	50	30	Citation	Mesh
Acer rubrum	Southeast	CWT	Broadleaf	×	×	×	×	(Ball and others 2009)	M
*Acer saccharum**	Northeast	BBRK	Broadleaf	×	×	×	×	(Rustad 1994)	M
Acer saccharum	Midwest	CDR	Broadleaf	×	×		×	(Hobbie 2005)	S
Acer saccharum	Southeast	CWT	Broadleaf	×		×	×	(Harmon and others 2009)	M
Acer saccharum	Northeast	HBR	Broadleaf	×	×	×	×	(Lovett and others 2016)	M
Acer saccharum	Northeast	HBR	Broadleaf	×		×	×	(Harmon and others 2009)	M
Acer saccharum	Northeast	HFR	Broadleaf	×		×	×	(Harmon and others 2009)	M
Betula alleghaniensis	Northeast	HBR	Broadleaf	×	×	×	×	(Lovett and others 2016)	M
Bromus inermis	Midwest	KBS	Grass	×	×	×	×	(Wickings and others 2012)	M
Cylindropuntia sp	Southwest	SRER	Cactus	×	×	×	×	Throop unpublished	M
Dacryodes excelsa	Tropics	LUQ	Broadleaf	×			×	Lodge unpublished	M
Digitaria californica	Southwest	SRER	Grass	×	×		×	Throop unpublished	M
Drypetes glauca	Tropics	GSF	Broadleaf	×		×	×	(Harmon and others 2009)	M
Drypetes glauca	Tropics	LUQ	Broadleaf	×			×	(Harmon and others 2009)	M
Drypetes glauca	Tropics	MTV	Broadleaf	×			×	(Harmon and others 2009)	M
Eragrostis lehmanniana	Southwest	SRER	Grass	×	×		×	(Throop and Archer 2007)	M
Fagus grandifolia	Northeast	BBRK	Broadleaf	×	×	×	×	(Rustad 1994)	M
Fagus grandifolia	Northeast	HBR	Broadleaf	×	×	×	×	(Lovett and others 2016)	M
Fraxinus americana	Northeast	HBR	Broadleaf	×	×	×	×	(Lovett and others 2016)	M
Isocoma tenuisecta	Southwest	SRER	Forb	×	×	×	×	Throop unpublished	M
Liriodendron tulipifera	Southeast	CWT	Broadleaf	×	×	×	×	(Ball and others 2009)	M
Miconia prasina	Tropics	LUQ	Broadleaf	×	×	×	×	(Prather and others 2018)	M
*Picea rubens**	Northeast	BBRK	Conifer	×	×	×	×	(Rustad 1994)	M
Piper glabrescens	Tropics	LUQ	Broadleaf	×		×	×	(Prather and others 2018)	M
Pinus resinosa	Northeast	HBR	Conifer	×		×	×	(Harmon and others 2009)	M
Pinus resinosa	Northeast	HFR	Conifer	×	×		×	(Harmon and others 2009)	M
Pinus strobus	Midwest	CDR	Conifer	×		×	×	(Hobbie 2005)	S
Prosopis velutina	Southwest	SRER	Broadleaf	×	×	×	×	(Throop and Archer 2007)	M
Quercus ellipsoidalis	Midwest	CDR	Broadleaf	×	×		×	(Hobbie 2005)	S
Quercus prinus	Southeast	CWT	Broadleaf	×		×	×	(Harmon and others 2009)	M
Quercus prinus	Northeast	HFR	Broadleaf	×		×	×	(Harmon and others 2009)	M
Quercus rubra	Southeast	CWT	Broadleaf	×	×	×	×	(Ball and others 2009)	M
Quercus spp.	Northeast	HFR	Broadleaf	×	×		×	(van Diepen and others 2015)	S
Rhododendron maximum	Southeast	CWT	Broadleaf	×	×	×	×	(Ball and others 2009)	M
*Trifoliumin carnatum**	Southeast	SVF	Forb	×		×	×	(Ball and others 2014)	S,L
Zea mays	Midwest	KBS	Grass	×	×	×	×	(Wickings and others 2012)	M

*Plant litter decomposition samples contributed to this study for which there were data available at both 100% and 30% dry mass remaining (and often 75% and/or 50%). Sites include Long-Term Ecological Research Sites [Cedar Creek (CDR), Coweeta (CWT), Harvard Forest (HFR), Hubbard Brook (HBR), Kellogg Biological Station (KBS), and Luquillo (LUQ)], as well as Bear Brook, Maine (BBRK), Guanica State Forest, Puerto Rico (GSF), Monte Verde, Costa Rica (MTV), Santa Rita Experimental Range, Arizona (SRER), and Spring Valley Farm, Georgia (SVF). Species followed by a *denotes cases where we collected data or samples from two separate ''projects'' at that same site using that species (for example, multiple mesh sized litterbags or native forests where it was decomposed). Mesh sizes are denoted by Small, Medium, and Large.*

in the southwest. Notably, several species were decomposed at multiple sites, in particular *Acer saccharum* in three regions and *Quercus prinus* in two regions. *Drypetes glauca*, *Fagus grandifolia*, and *Pinus resinosa* were decomposed at multiple sites within just one region.

In addition to these collected samples, we include data from published studies where (1) data were included for the same criteria of 100% and 30% mass remaining, and (2) chemical data were available at those stages for at least some of the parameters we analyzed following the same method of chemical analysis. This allowed us to further incorporate data from a subset of species from LIDET, as well as from another study of four species.

Data Analyses

All statistical analyses were conducted in R (version 4.0.2, The R Foundation). Litter chemistry parameters were analyzed by first calculating a distance matrix using the ''vegdist'' function in the package ''vegan''. This was done separately for (1) percent content of major nutrients (N, P, K, Ca, Mg), (2) percent content of C and C-based fiber compound categories determined by sequential acid digestion (NDS, ADS, AS, AI) abbreviated in tables as C + Fiber, and (3) percent content of molecular classes determined by py-GCMS (lignin, polysaccharides, phenols, lipids, N-bearing, proteins, aromatic, and the unknown category). Note that py-GCMS molecular classes are analyzed separately from the other structural compound chemistries because the data can only be expressed as percent content relative to all measured compounds, not absolute mass per g litter, and is therefore not numerically compatible with the other chemical parameters. A permutation multivariate analysis of variance (PERMANOVA) was then performed on the distance scores using the ''adonis'' function to determine the effects of decay stage, region, and functional type of the litter (including their interactions) on overall litter chemistry. When main effects were significant, a pair-wise PERMANOVA was conducted for that main effect using the ''pairwise.perm.manova'' function of package ''RVAideMemoire''. Where interactions of region or functional type with stage were significant, a separate PERMANOVA was run on each stage individually. Data were visualized using a non-metric multidimensional scaling (NMDS) using the function ''metaMDS'' in package ''vegan''.

Following up from this PERMANOVA, individual litter chemistry parameters were analyzed in a three-way Analysis of Variance (ANOVA) testing for the effect of decay stage, region, and functional type, as well as their interactions, on each parameter. Doing so allowed us to describe the way in which functional type and geographic region influences that parameter, and whether it changes over time through an interaction with stage. If region or functional type were significant, it was followed by a post-hoc Tukey test to show which ecosystems differed significantly from each other. To meet the assumptions of normality and heteroscedasticity, most parameters had to be log-transformed: all nutrient elements (excluding carbon), all nutrient ratios, and all compound classes except polysaccharides and lignin.

Results

Broad-Scale Patterns of Litter Chemistry across Decay Environments and Plant Functional Types

The PERMANOVA of the three chemical categories (nutrients, C + fiber, and molecular classes identified by py-GCMS) showed that, when looking across all of the samples collected, litter chemistry significantly differed according to decay stage, geographic region of decay, and plant functional type (Table 2). However, the relative importance of those three main effects differed among the chemical categories. Functional type, followed by decay stage, explained the largest portion of the variability in nutrient chemistry (denoted by their R^2), with only a small fraction of the variability explained by region and interactions among main effects. For C-based fiber chemistry, the greatest explanation of variability was again functional type, followed by stage and region fairly equally, and interactions explained much less of the variability. However, it was geographic region that had the largest influence on molecular classes measured by py-GCMS, followed by functional type to a lesser degree; decay stage was not significant. Thus, the functional type of litter played a bigger role in structuring nutrient and C-based chemistry during decomposition than did the environment in which it's decomposing and stage of decay, but the environment in which it's decomposing played the bigger role in molecular class chemistry. Overall, though, only about three quarters of the variability in each chemical category was explained by stage, region, and functional type.

Despite the fact that region and functional type differed in relative importance for each chemical category, they were both significant factors for all three categories. Pairwise comparisons demonstrate that relationships among the regions (midwest, southwest, northeast, southeast, and tropics) and functional types (broadleaf woody plants, coniferous woody plants, cacti, grass, and forbs) differ across the three chemical categories (Table 2, Figure 1). Among the plant functional types, cacti were characteristically high in Mg and Ca, grass high in P but low in Mg and Ca, and conifers low in both macro- and micronutrients, while forbs and broadleaf plants contained a similarly broad nutrient makeup (Figure 1a). For fiber compounds, cacti and forbs (low in structural materials and higher in NDS-compounds), grass (high in ADS- and AS-compounds but low in AI-compounds), and broadleaf (high in AI-compounds but moderate in

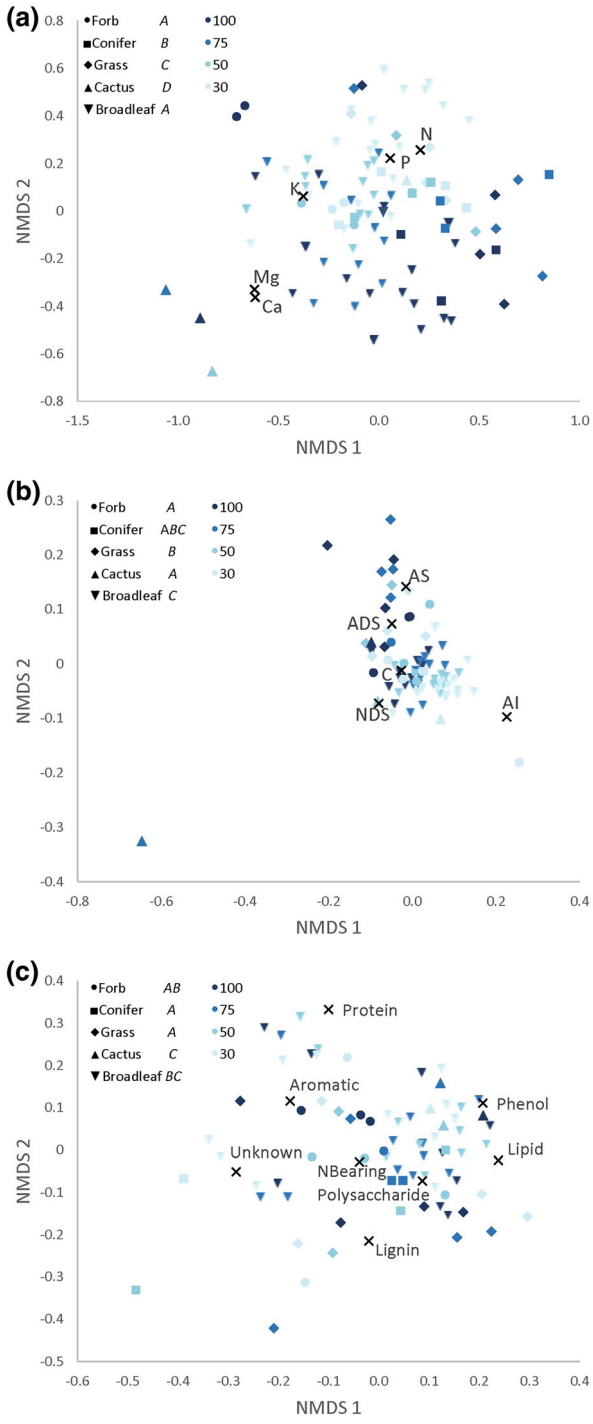
Table 2. *P*-Values from the PERMANOVA on Chemical Categories.

	Nutrients		C + Fiber		py-GCMS	
	R^2	*P*	R^2	*P*	R^2	*P*
All Data						
Stage	0.163	**0.001**	0.106	**0.001**	0.010	0.137
Region	0.090	**0.001**	0.187	**0.001**	0.424	**0.001**
Functional type	0.254	**0.001**	0.251	**0.001**	0.147	**0.001**
S*R	0.057	**0.001**	0.042	**0.022**	0.037	0.088
S*F	0.042	**0.002**	0.060	**0.003**	0.018	0.640
R*F	0.056	**0.001**	0.031	**0.008**	0.051	**0.001**
S*R*F	0.011	0.225	0.046	**0.001**	0.021	0.093
Residuals	0.326		0.278		0.292	
100%						
Region	0.286	**0.001**	0.222	0.072	0.454	**0.016**
Functional type	0.384	**0.001**	0.483	**0.002**	0.235	**0.043**
R*F	0.057	0.053	0.048	0.171		
Residuals	0.273		0.247		0.311	
75%						
Region	0.084	0.427	0.288	**0.009**	0.388	**0.002**
Functional type	0.525	**0.001**	0.563	**0.001**	0.268	**0.039**
R*F	0.075	0.074	0.015	0.406	0.043	0.263
Residuals	0.316		0.132		0.300	
50%						
Region	0.264	**0.048**	0.334	**0.001**	0.478	**0.004**
Functional type	0.458	**0.002**	0.516	**0.001**	0.201	0.192
R*F			0.051	**0.024**		
Residuals	0.279		0.099		0.321	
30%						
Region	0.214	**0.025**	0.316	**0.001**	0.446	**0.003**
Functional type	0.328	**0.001**	0.245	**0.004**	0.158	0.146
R*F	0.015	0.907	0.167	**0.002**	0.062	0.138
Residuals	0.443		0.272		0.333	

Resulting P-values from the PERMANOVA testing the impacts of decay Stage, geographic Region, and litter Functional type on litter nutrient, metabolic, and structural chemistry across all sampling periods and individually for each stage bin of mass remaining. Significant interactions ($P < 0.05$) are in bold to add emphasis.

ADS-compounds) litter were all distinct from each other, with conifer litter similar to all (Figure 1b). For molecular classes, cacti and broadleaf (low lignin but high in proteins and lipids) grouped separately from grass and coniferous trees (high lignin but low in proteins and lipids), with forbs intermediate between the groups (Figure 1c).

The distinctions among regions also depended upon the chemical categories under consideration. Litter decomposed in the northeast was lower in most nutrient elements than that from the tropics and southeast, with all other regions intermediate between those two (Appendix 1a). There were three distinct regional groupings of C-based fiber chemistry: tropical litter that was low in C and ADS-compounds; eastern (northeast and southeast) litter that was high in C and AI-compounds; and western (midwest and southwest) litter that was low in AI-compounds but high in ADS-compounds (Appendix 1b). For the molecular classes, all regions were distinct, except for similarities of the southeast with the northeast and midwest, which notably share broadleaf species (particularly *Acer saccharum*) and encompass the only agricultural sites (southeast and midwest) (Appendix 1c). Litter differentiates between the northeast (high phenols and lipids), southwest (high in lipids and polysaccharides), midwest (moderately high in lignin, N-bearing, and compounds of unknown origin), and tropics (high in proteins and aromatics). Notably, these patterns appear true across all of functional types present within each region, with the exception of grass driving the differentiation of western litter in their C + fiber and polysaccharide characteristics and the other functional types behaving more similarly to other regions (compare Figure 1b with Appendix 1b). Beyond this grass influence, the patterns described here hold true



◀**Figure 1.** NMDS ordinations of three categories of leaf litter chemistry: **a** nutrients with stress 0.126; **b** carbon and carbon-based fiber compounds determined by sequential acid digestion with stress 0.111; and **c** structural and metabolic compounds determined by py-GCMS with stress 0.186. Data points represent the average value for the replicates of each project ($n = 4$) and are organized according to plant functional type of the litter and stage of decay. Italicized letters demonstrate significant pairwise comparisons, where functional types with the same letter do not differentiate from each other.

even when considering only broadleaf species, which was the only functional type decomposed in all regions (Appendix 2).

The influence of decay stage on nutrient and C-based fiber chemistry demonstrates the trajectory of litter chemistry throughout decay as it varies across geographic regions and plant functional types (Figure 1a, b). Even when viewed across regions, broadleaf, conifer, and (to an extent) grass litter tended to increase in N and P over decay, but notably forbs and cacti did not. All functional types tended to increase in AI compounds (commonly associated with lignin) as AS compounds decline later in decay. With the lack of effect of decay stage, molecular classes do not follow a notable trajectory throughout decay (Figure 1c).

Region and functional type significantly interacted with decay stage for nutrients and carbon-based fiber chemistry, but they did not for molecular class chemistry (Table 2). Thus, the nature of the regional and functional type influences on molecular classes persisted throughout the first 70% of mass loss, but varied over time for nutrient and fiber classes. When the PERMANOVA was run separately for each stage bin, distinctions among functional types in their nutrient chemistry persisted throughout the decay stages (Table 2), though the nature of the relationships shifted slightly throughout time (Figure 1a). In other words, litter from across functional types did not totally converge on similar nutrient chemistry across the first 70% of decomposition, but instead maintained distinct nutrient chemistry. Regions also maintained distinctions in their nutrient chemistry, though the size of those differences declined a bit over time (Table 2). This was largely driven by the fact that the initial difference of northeast litter due to its low nutrient content diminished by 75% mass remaining. For C-based fiber chemistry, initially only functional type was significant, largely influenced by a significant difference between broadleaf and grass that persists

Table 3. *P*-Values from the Three-Way ANOVA on Each Litter Chemistry Parameter

	Stage	Region	Functional type	S*R	S*F	R*F	S*R*F
Nutrient chemistry							
N	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.002**	< **0.001**	< **0.001**
P	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.032**	**0.018**	0.474
C:N	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.691
C:P	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.001**	< **0.001**	0.556
N:P	**0.005**	< **0.001**	< **0.001**	**0.002**	0.068	0.073	0.217
Ca	**0.011**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.333	0.092
K	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.001**	0.157
Mg	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.860	0.052
Na	**0.020**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.671
Carbon and fiber (SAD)							
C	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.006**	< **0.001**	< **0.001**
AI	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.065	**0.004**
AS	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.138	**0.005**
ADS	**0.020**	< **0.001**	< **0.001**	0.278	**0.003**	0.773	0.406
Metabolic and structural compounds (py-GCMS)							
Aromatic	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.033**	0.264
Lignin	< **0.001**	< **0.001**	**0.001**	**0.020**	0.292	0.785	0.297
N-Bearing	**0.047**	**0.014**	< **0.001**	< **0.001**	**0.013**	< **0.001**	0.392
Proteins	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.794
Lipids	0.111	< **0.001**	< **0.001**	**0.002**	**0.002**	< **0.001**	0.338
Phenols	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	0.129
Polysaccharides	**0.001**	< **0.001**	< **0.001**	0.327	0.599	0.088	0.752
Unknown	< **0.001**	< **0.001**	< **0.001**	< **0.001**	< **0.001**	**0.001**	0.143

Resulting P-values from the three-way ANOVA performed on each litter chemistry parameter showing the effect of decay Stage, geographic Region, and plant Functional type. Significant interactions ($P < 0.05$) are in bold to add emphasis.

throughout decay. At 75% mass remaining and beyond, region was also a significant factor. Overall, there is little evidence that litter chemistry from across a broad spectrum of ecosystems and functional types converged on a homogenous litter chemistry across the board. Litter functional types remained distinct for all chemical categories (through 30% mass remaining, at least), and regional differences diverged slightly (for C + fiber), remained distinct but shifted over time (nutrients), or remained the same (py-GCMS molecular classes) throughout decay.

Additionally, region significantly interacts with functional type for all three chemical categories (Table 2). This suggests that the nature of the persistent influence of plant litter functional type on chemistry differs across the regions. In other words, the influence of region on litter chemistry during decomposition is not consistent across all functional types. Because of the unbalanced replication and the fact that not all functional groups were decomposed in all regions, the nature of this interaction is difficult to statistically assess.

Response of Individual Chemical Parameters to Stage, Climate, and Functional Type

Given that region and functional type significantly influenced all three chemical categories, we also explored the behavior of individual chemical parameters throughout the decay stages (Table 3). Each chemical parameter differed significantly according to region and functional type, and all except lipids significantly changed over decay stages.

A significant interaction of region or functional type with decay stage would indicate that the nature of their influence on chemical parameters changes during decomposition. We found significant interactions of region with decay stage for all parameters except ADS compounds (including hemicellulose) and polysaccharides (Table 3). Functional type significantly interacted with decay stage for all of the parameters except N:P, lignin, and polysaccharides. Where the interaction was significant, we investigated the direction of change to identify possible convergence or divergence over time (Figures 2 and 3, Appendices 3 & 4). Some

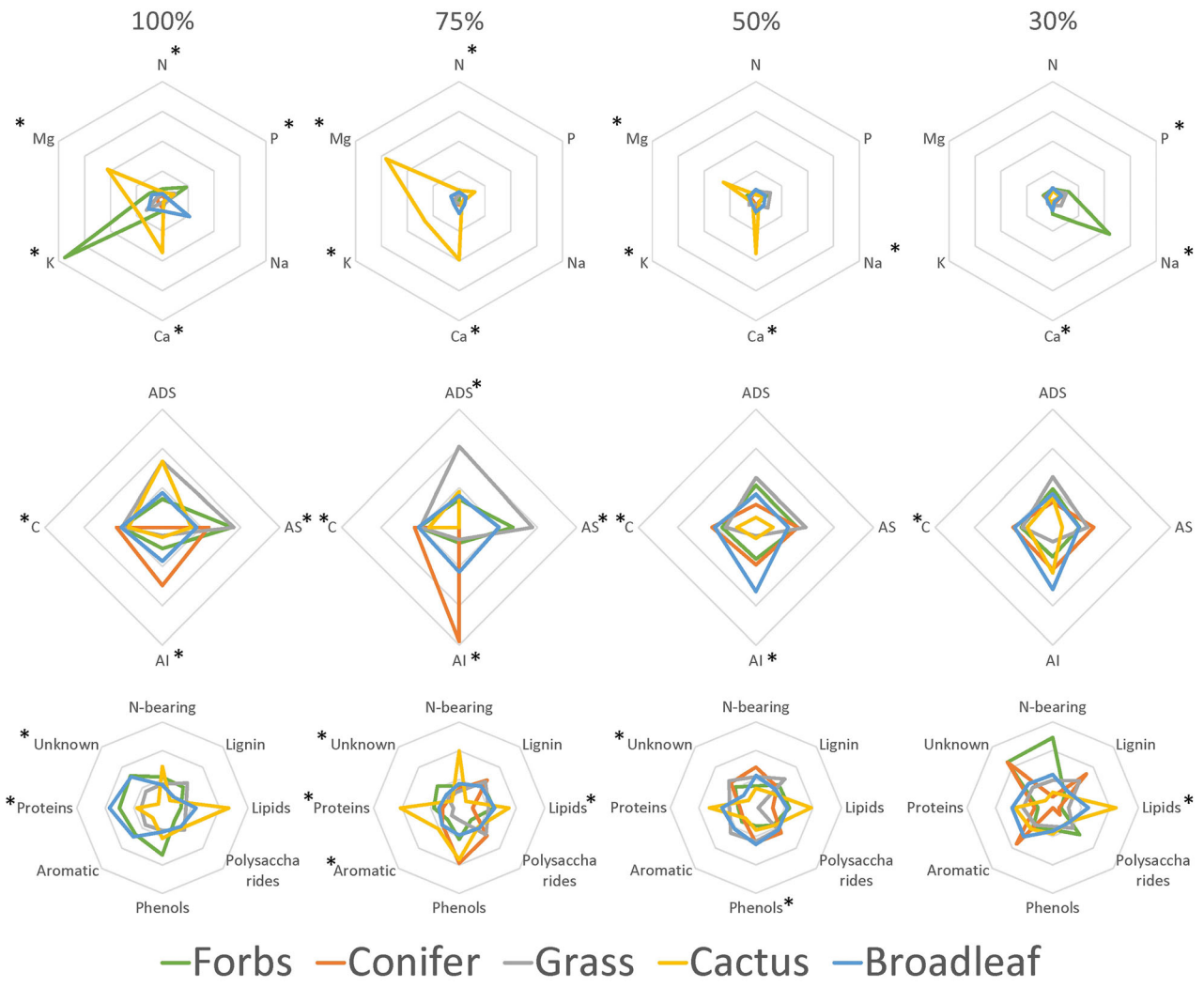


Figure 2. Radar plots demonstrating the differences in litter chemistry among functional types at different decay stages. Parameters for which there is a significant difference among the functional types at that decay stage are denoted with an asterisk (*). For example, functional types differed significantly from each other in their %N content at 100% mass remaining, denoted by the * next to the N axis and viewed as a separation of the lines as they cross the N sector. However, they do not significantly differ at 30% remaining (no * next to the N axis) where the lines overlap at this point, suggesting convergence. Note that data were standardized as a proportion of the mean value for each chemical parameter, to allow multiple parameters to be plotted on the same scale. Actual values for each point, and associated statistics, can be found in Appendix 3.

chemical parameters were initially different among functional types (N, K, Mg, AS compounds, proteins, and unknown compounds; Figure 2) and regions (AS compounds; Figure 3) to then lose that statistical distinction, in some cases even converging on similar chemistry across the groups later in decomposition. For some parameters, the lack of distinction was due to lower replication reducing statistical power to detect differences (for example, the structural & metabolic compounds), while in others it appears to be genuine convergence on a homogenous chemistry (nutrients N, K, Mg in Figure 2). Alternatively, others were initially similar across regions (C, N-bearing, phenols) and functional types (Na, lipids) but diverged to become distinct later in decomposition. The majority of chemical parameters, however, neither diverged nor converged, with differences among regions and functional groups persisting throughout decay.

Overall, regional differences in litter chemistry were largely maintained throughout decay (neither diverging nor converging; Figure 3). This is particularly true for nutrients, nutrient ratios, and many molecular classes, though the nature of regional

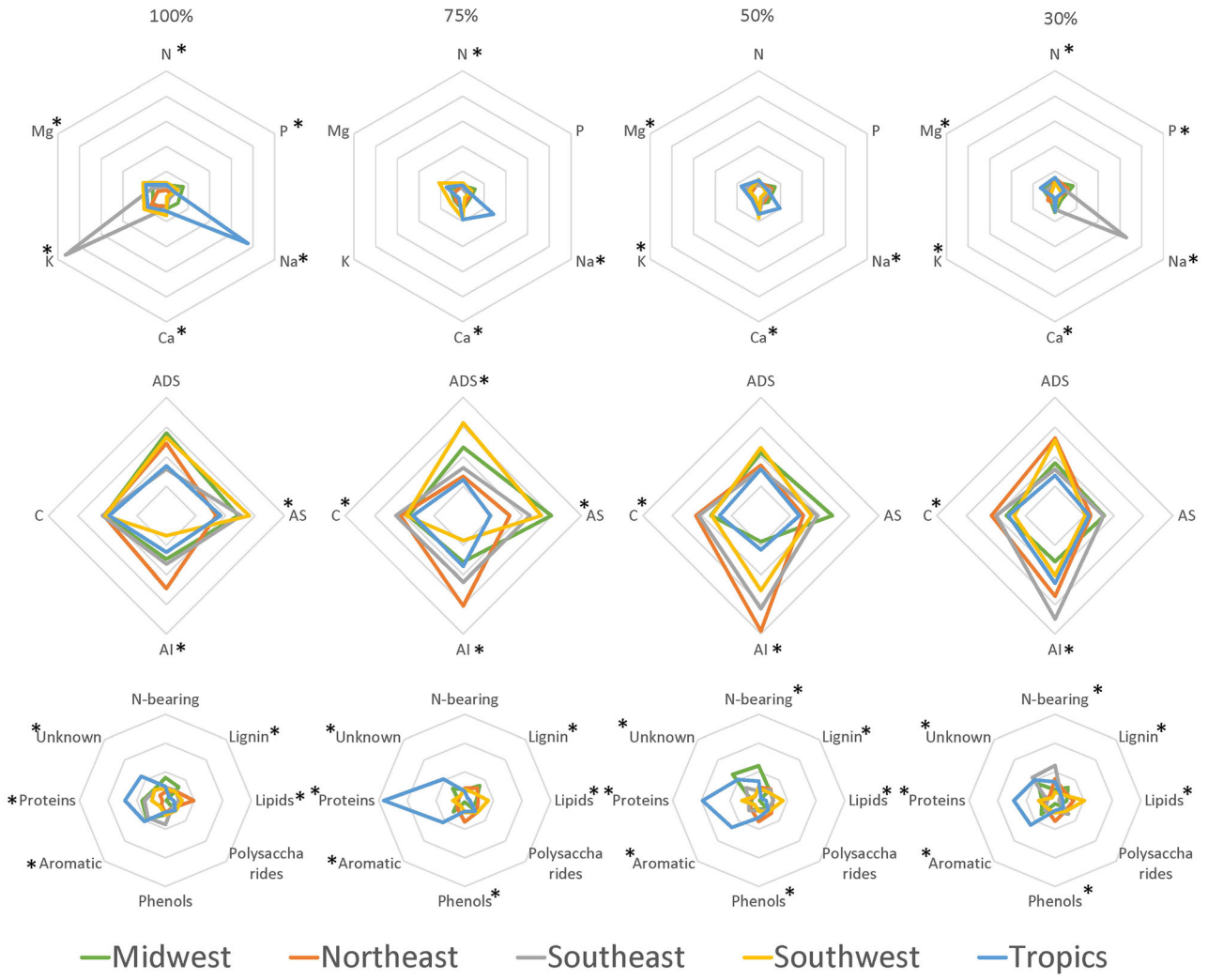


Figure 3. Radar plots demonstrating the differences in litter chemistry among regions at different decay stages. Parameters for which there is a significant difference among the regions are denoted with an asterisk (*). For example, regions differed significantly from each other in their %C content at 30% mass remaining, denoted by the * next to the C axis and viewed as a separation of the lines as they cross this point. However they did not significantly differ at 100% remaining (no * next to the C label) where the lines overlap as they cross this point, suggesting divergence over time. Note that data were standardized as a proportion of the mean value for each chemical parameter, to allow multiple parameters that differ greatly in magnitude to be plotted on the same scale. Actual values for each point, and associated statistics, can be found in Appendix 4.

differences in many nutrients changed over time (N, P, K, Na, Ca, C:N, C:P, as well as AI compounds). Molecular class chemistry largely maintained initial differences that did not shift over time, with only a few instances of modest divergence over time. Functional types, however, demonstrated several instances of chemical homogenization for several nutrients (Figure 2). Most other nutrients maintained initial differences, as they did among regions. Evidence of divergence was limited in functional types, with only modest divergence detected in Na and lipids. The convergence of some nutrients among functional types while regional differences are maintained reflected that the trajectory they were following over time toward functional convergence differed across regions, causing the nature of the regional differences to shift over decay stages.

Region also significantly interacted with functional type for many of the parameters, including C, N, P (and their ratios), K, Na, and most of the molecular classes from py-GCMS (Table 3), suggesting that the chemical differences among plant functional types are sensitive to the region in

which they are decomposed. This interaction was most notable in broadleaf trees, which is the only functional type for which we have data in all of the regions (Appendix 5). Chemical content of broadleaf species differed among regions in all nutrients, C, AI compounds, and most py-GCMS compound categories. This interaction also means that differences among functional types were not uniform across the regions, reflecting the differences among species found within those functional types. For example, in the southwest, a legume (mesquite) caused broadleaf litter to contain higher nutrients compared to forbs, while the reverse was true in the southeast where a legume (clover) caused the forbs to have higher nutrients than the broadleaf taxa.

Discussion

The goals of this research were to identify (1) common patterns of litter chemical changes throughout decay, such as chemical convergence or divergence, across geographic regions and plant functional types, and (2) whether these patterns are consistent across litter chemical parameters. While our results highlight the importance of the ecosystem's characteristics (summarized by different geographic regions) and plant functional type in driving litter chemical changes throughout decomposition, the short answer to these questions is that we do not find universally common patterns of litter chemical trajectories across all functional types and regions, and very limited evidence of convergence or divergence in chemistry over time, mostly within the nutrient elements. The established understanding of chemical changes in the published literature, with immobilization of nutrients and concomitant increase of recalcitrant compounds, seems to be true across regions for the commonly-studied functional types (woody broadleaf and conifer taxa and grass), although it is not necessarily true for other taxa (forbs and cacti). Therefore, regions that are dominated by woody species or grass seem likely to follow these familiar patterns across functional groups, but ecosystems where forbs and cacti dominate appear to be less predictable over time. Further work that expands the number of species within these broadly-defined functional groups will be necessary to determine whether the patterns are true, but our data suggest large implications for the predictability of decomposition if the changes in the dominant vegetation across ecosystems resulting from human-induced global environmental change (Franklin and others 2016; Komatsu and others 2019) and the expansion of arid ecosystems (UNDP 2007) result in a shift among these functional types.

Broadly speaking, functional types had a particularly strong impact on nutrient and fiber chemistry, but a secondary influence on molecular class chemistry. Although functional types converged over time for a few of the nutrient elements (N, K, Mg), with marginal divergence of Na and lipids, the overarching pattern was for persistence of distinct elemental and compound chemistry. The influence of geographic regions on nutrient and fiber chemistry was secondary to functional type, but it had a dominant control over molecular classes. Interestingly, chemical change in molecular classes responded to the decomposition environment more strongly than the nutrients and fiber chemistry that are more frequently measured in decomposition experiments. This perhaps reflects a greater influence of microbial evolutionary adaptation on the processing of the metabolic chemical classes measured by py-GCMS, wherein breakdown of particular classes (lipids, proteins and other N-bearing compounds, phenols and other aromatic compounds) is more sensitive to environmental factors of its 'home field' ecosystem (similar to the findings on homefield advantage for litter chemistry by Wallenstein and others 2013) than it is to physiological differences in compound availability among functional types. The dominant influence of region, that again persists over time (with only a few instances of divergence) over these compounds suggests that the trajectory of chemical change during decay will be particularly sensitive to climate change. Methodology for measuring chemical classes based on molecular structures is relatively new technology compared to the long-standing techniques for C and macronutrients, and as their measurement becomes more common in litter decay studies, it will become clear how universal this overarching pattern is.

Thus, in sum, our data suggest that, when thinking broadly across taxa and ecosystems, there are limited instances of convergence, largely in nutrient chemistry, that occur across functional types but not regions. Divergence occurred to a limited extent in nutrients and chemical classes, both across functional types and regions. Even then, these examples of convergence and divergence result from the specific species collected in this study, and could potentially be nullified with the addition of more species to each functional group. Overall, different ecosystems tend to maintain unique chemical trajectories throughout decay, though sometimes functional types within an ecosystem will converge on similar nutrient

chemistry. This illustrates that the instances of convergence and divergence that have been reported in the published literature (Wickings and others 2012; Liu and others 2016; Wang and others 2019; Du and others 2020) are specific to the litter species and chemical parameters studied, and when compiled across taxa and ecosystems do not comprise a broad-scale pattern of convergence or divergence.

Although the overarching persistence of chemical differences is generally consistent across ecosystems and functional types, the nature of the relationships among regions or litter types often shifted throughout decay. For example, for most of the parameters in Figure 3, initial differences among regions (stage bin 100) are not the same as the differences among the exact same litter species per region at stage bin 30. This explains why regional differences in litter nutrients overall became less distinct over time in the PERMANOVA analysis despite a lack of convergence in individual nutrient parameters; it is not chemical convergence on a homogenous nutrient chemistry across regions, but a lack of distinction at some time points due to variability in individual trajectories of nutrient mobilization and immobilization.

Our results also demonstrate that we cannot consistently predict one typical trajectory of chemical change that is true across all ecosystems and plant functional types. This is due in part to the different individual species in each functional group that are native to the different regions. For example, the northeast, southeast, and tropics are all broad-leaf dominated, yet C + Fiber chemistry in the southeast and northeast (who share many similar species of broadleaf trees) group together separately from tropics where the broadleaf trees are entirely different species. The northeast has lower nutrient content, due to both broadleaf species (including shared species with the SE) and the number of low-nutrient conifer studies in the northeast compared to the southeast and tropics. The midwest and southwest group together in C + Fiber chemistry, reflecting the prominence of grass species in these ecosystems with characteristically different AI and ADS content. Yet, in molecular classes, regions all differ despite similarities among the functional groups represented. Regardless of the different native species found in each region, the Region*Function interaction still shows a lack of consistency at broad scales, suggesting that no general patterns about litter chemical trajectories within a particular functional group studied within one region will be true in others. The lack of a consistent pattern across regions is also identified in individual cross-site studies in the literature. A gradient of four European sites also identified an interaction of species identity with forest type to influence mass loss, suggesting that litter chemical differences depended upon site-specific conditions (Zhou and others 2020). The rate at which sugar maple, one of the key species for which we collected litter from across regions, lost mass and nutrients in different forests of Quebec varied without a distinctive pattern, independent of site and forest type (Bélanger and others 2019). Our data supports these regional-scale findings with continental-scale data demonstrating independent chemical trajectories among sites.

Decomposition of litter, including its many chemical components, ultimately contributes to C and nutrient cycling in soils. Given the lack of predictable homogenization of litter chemistry, it is difficult to predict the rates and patterns of chemical breakdown and nutrient release, at least during the first 70% of mass loss across functional types and regions. If litter functional type (that is, unique litter chemistry) is a primary driver of decomposition and decay environment (for example, climate) is secondary, understanding shifts in vegetation communities is key to understanding how climate change will indirectly influence the flux of C and nutrients from litter to soil. Because each functional type can have unique chemical trajectories without overall homogenization in chemistry, vegetation shifts that result in functional type replacements will impact the trajectory of litter chemistry that impacts C storage and soil fertility. Additionally, because the byproducts of these compounds are incorporated into the soil as a basal resource for soil biota and are ultimately incorporated into microbial biomass and soil organic matter (SOM), future work should investigate their importance at different stages of litter decay for food web dynamics and SOM formation (Joly and others 2020).

Our results also highlight the benefit of long-term sample and data archives, such as are made available by Long-Term Ecological Research (LTER) networks. Such networks provide the foundation for cross-ecosystem comparisons of similar plant functional types across multiple climates, soil types, and biotic communities over long periods of time that are required for understanding decomposition dynamics and SOM formation. Future cross-site work should further explore how these chemical changes throughout decomposition relate to mass loss and therefore C dynamics. For example, it is still unclear how tightly these various chemical

parameters parallel C and overall mass loss, and whether knowledge of litter chemistry throughout decay significantly improves predictions of mass loss across this broad range of functional types and ecosystems.

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