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Fog as a source of nitrogen for redwood trees: evidence from fluxes and stable isotopes

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Summary

1. A defining feature of the redwood forest in coastal California is the presence of fog in the summer months, a time when there is typically little rainfall. Our goal was to determine the role of summer fog in canopy transformation of nitrogen, nitrogen uptake by trees and photosynthesis within a coastal redwood forest ecosystem.
2. We measured horizontal and vertical inputs of nitrogen, the isotopic composition of nitrogen in a variety of atmospheric sources (summer fog, winter rain and throughfall throughout the year), nitrogen pools (soil solution) and plant tissue (roots and foliage), as well as rates of photosynthesis and nitrogen uptake by trees.
3. Throughfall nitrogen fluxes were greater at the forest edge compared to the interior both within the canopy (sampled 10 m above-ground) and onto the forest floor (sampled 1 m above-ground; $P < 0.05$). Similarly, soil solution NO_3^- and total inorganic nitrogen were greater at the forest edge compared to the interior ($P = 0.0014$ and 0.009, respectively). Whereas natural abundance $\delta^{15}NO_3$ values were not significantly different between winter rain (measured as bulk precipitation) and summer fog water (average $\delta^{15}N = -1.2 \pm 0.68^0/_{00}$), $\delta^{15}NH_4$ values were significantly greater in fog water ($11.4 \pm 2.7^0/_{00}$) compared to rain ($1.2 \pm 0.9^0/_{00}$). We found no difference in $\delta^{15}N$ in roots from forest edge trees compared to interior trees. In contrast, nitrogen concentrations and $\delta^{15}N$ in foliage from forest edge trees were significantly greater compared to interior trees ($P < 0.0001$), suggesting that the leaves of forest edge trees may be obtaining a greater proportion of their nitrogen from fog compared to those of the interior trees. Natural abundance ^{13}C of leaf sugars and rates of photosynthesis were significantly higher at the forest edge compared to the interior during the fog season ($P < 0.05$), but not different between locations in the rain season ($P > 0.05$). Nitrification in the

forest floor, rather than the canopy, is the primary source of NO_3^- in these soils throughout the year.

4. *Synthesis*. Summer fog provides nitrogen directly and indirectly to redwood trees, especially those at the forest edge, and affects the physiologic function of redwood trees.

Introduction

Fog provides significant water and nutrient inputs to both minimally disturbed (Weathers & Likens 1997; Weathers *et al*. 2000; Ewing *et al*. 2009; Barbosa *et al*. 2010; Hiatt, Fernandez & Potter 2012) and polluted ecosystems (Kimball *et al*. 1988; Igawa *et al*. 1998; Fenn *et al*. 2000). Summer fog in Mediterranean habitats is produced from the mixing of warm, high-pressure air and cold, upwelled water within the nearby coastal ocean that are then carried inland via wind (Petterssen 1936). Similar to other Mediterranean habitats, coastal redwood (*Sequoia sempervirens*) forests receive fog water in the summer months at a time when the soils would otherwise be dry and the vegetation would experience water deficits since < 3% of annual precipitation is delivered during the summer months (Ewing *et al*. 2009; Hiatt, Fernandez & Potter 2012). Coast redwood trees are the tallest on Earth and, today, are constrained to a narrow belt along the coast of northern California and southern Oregon (Noss 2000). Uptake of fog water by redwood trees during the summer months represents up to 45% of the source of water for annual transpiration (Dawson 1998).

In fog-inundated ecosystems, there has been increased attention to linkages between water and nitrogen availability (González *et al*. 2011) since both are essential resources for trees and the understorey plants that inhabit these ecosystems. Some studies in European forests and in high-elevation spruce forests of the USA have demonstrated direct foliar uptake of nitrogen, although rates are typically quite small compared to root uptake (Bowden, Geballe & Bowden 1989; Harrison *et al*. 2000; Sparks 2009). Across a range of redwood sites in California, foliar concentrations of nitrogen have been shown to be greater in wetter (more northern) sites, although it is unclear whether this pattern is due to microbial processes that result in greater nitrogen availability in soils or from greater atmospheric nitrogen inputs (Santiago & Dawson 2014). Similar to other fog-inundated ecosystems around the world (Weathers *et al*. 1988; Weathers & Likens 1997; Weathers *et al*. 2000; González *et al*. 2011), concentrations of nitrogen in the fog of redwood forests are significantly greater than those in rainfall. In fact, nitrogen from canopy throughfall during the summer fog season in redwood forests provides up to 21% of annual nitrogen inputs to the forest floor (Ewing *et al*. 2009). Due to the horizontal flow of fog, redwood trees at the coastal edge of stands can get up to seven times more water and nitrogen via throughfall (drip) to the forest floor compared to interior trees (Ewing *et al*. 2009). In contrast, rain falls primarily vertically, creating less of a contrast in water and nitrogen inputs as a function of location within a forest stand.

Rates of gross nitrification in soils of redwood forests have been found to correlate positively with moisture availability (Bradbury & Firestone 2007), but we are unaware of any studies examining the potential role of nitrification in redwood forest canopies. Redwood trees, especially those in their northern range, are unique in having some organic soil within the canopy (Enloe, Graham & Sillett 2006), which could contribute to biogeochemical cycling of nitrogen within these ecosystems. Previous work in cloud forests of Costa Rica shows that nitrogen cycling within arboreal mats can represent significant fluxes of nitrogen that are independent of soils on the ground (Hietz *et al*. 2002; Clark *et al*. 1998). While microbial transformation of nitrogen within the canopy of redwood forests is not likely to be as large in magnitude as microbial processing in ground-based soils, nitrogen entering the redwood canopy in summer via fog may be transformed *in situ* through microbial processes such as nitrification.

Numerous studies have taken advantage of distinct ^{15}N signatures of nitrogen sources to partition the relative contribution of those sources for plant nitrogen status (Dawson *et al*. 2002). For example, natural abundance ^{15}N values of NH_4^+ and NO_3^- have been used in ecosystem studies to examine nitrogen emissions from livestock waste, coal-fired power plants and tailpipe exhaust, since these sources of nitrogen vary in their ^{15}N isotopic signatures (Moore 1977; Heaton 1990; Elliott *et al*. 2007; Felix *et al*. 2013; Hastings, Casciotti & Elliott 2013; Beyn, Matthias & Dahnke 2014). Evans and Ehleringer (1974) examined natural abundance ^{15}N in plants along a fog gradient in the Atacama Desert, Chile, and did not find a significant difference in natural abundance ^{15}N between fog-inundated and non-inundated areas, concluding that these plants rely on infrequent rain events rather than fog as a source of nitrogen. In contrast, more recently, González *et al*. (2011) demonstrated the importance of fog nutrient (nitrogen as well as phosphorous) inputs to bromeliad stoichiometry in the Atacama Desert. Santiago & Dawson (2014) found a positive relationship between water inputs and foliar nitrogen concentrations of understorey plants across three redwood forests in California, but a negative relationship between total water inputs and foliar ^{15}N abundance. The greater amount of ^{15}N in foliage of understorey plants at the drier sites could indicate a shift from rain to fog water as a primary source of nitrogen since fog water can be more enriched in ^{15}N than rainwater (Templer *et al*. 2006). Together, these results suggest that understorey plants living in redwood forest sites with greater water availability may have greater nitrogen uptake. However, direct rates of nitrogen uptake by understorey plants were not measured in this study so it is unclear whether patterns of natural abundance ^{15}N in foliage are due to differences in rates of nitrogen uptake or in soil available nitrogen.

Because it has been shown that fog water provides significant amounts of both moisture and nitrogen to the forest floor during the otherwise dry summer season and these inputs move into the forest primarily horizontally, we quantified nitrogen inputs (fog and rain) and characterized their isotopic

composition across seasons (fog vs. rain) and forest location (forest edge vs. interior; canopy vs. forest floor) in a coastal redwood forest of California to assess whether and how fog nitrogen might affect plant and ecosystem processes. We predicted that within the windward, western-facing, most fog-inundated edge of the coastal redwood forest, foliage and roots would have (i) higher nitrogen concentrations than interior forest trees and (ii) natural abundance ^{15}N values that are more similar to fog than rain inputs due to greater nitrogen uptake from fog. We also predicted that (iii) nitrification within the canopy transforms some NH_4^+ deposited by summer fog to NO_3^-, which then may be delivered to the forest floor. We measured horizontal and vertical inputs of nitrogen, the isotopic composition of nitrogen in a variety of atmospheric sources (summer fog, winter rain and throughfall throughout the year), nitrogen pools (soil solution) and plant tissue (roots and foliage), as well as rates of photosynthesis and nitrogen uptake by trees, to increase our understanding of the movement and transformation of nitrogen within fog-inundated coastal redwood forest ecosystems. We estimated the relative contributions of precipitation (bulk precipitation in winter and fog water in summer) vs. nitrification to NO_3^- pools in the canopy and forest floor using the natural abundance isotope composition of NO_3^- in a two-end-member mixing model (Pardo *et al*. 2004).

Materials and methods

Study site

This research was conducted in an 11 ha remnant old-growth 400+-year-old redwood forest located on a ridge at 300 m elevation, in Sonoma County, California (38º 24′N, 122 º 59′W), approximately 8 km from the ocean (Burgess & Dawson 2004). The climate is Mediterranean with warm dry summers (mean annual July temperature, 19.6 °C for 1971–2000, National Weather Service, Santa Rosa, CA, 20 km north-west of the site) and rainy, mild winters (mean annual January temperature, 8.9 °C). More than 97% of the annual rainfall (mean annual equal to 78 cm, National Weather Service, Santa Rosa mean 1971–2000) typically falls between October and May (hereafter rain season). Soils in this forest are composed primarily of fine sandy loams of the Goldridge series (Ultisols, USDA 1972), with A horizons ranging from 60 cm at the forest edge to 11 cm at the most interior site (Ewing *et al*. 2012). The landscape adjacent to the remnant forest is comprised of grassland, agriculture (grapes), deciduous forest, second-growth redwood forests and scattered residential development.

Nitrogen in throughfall

We took advantage of throughfall samples collected from June 2004 through April 2005 at this site to measure natural abundance ^{15}N in NH_4^+ and NO_3^- in throughfall (see Ewing *et al*. 2009 for details of sampling, as well as nitrogen concentrations and fluxes). Briefly, we sampled throughfall from an array of 44 throughfall collectors distributed throughout the forest in a stratified

random design following Weathers, Lovett & Likens (1995, Willard *et al*. 2001) and described in detail in Ewing *et al*. (2009). Eight collectors were placed randomly within each of five ~60-m-wide north–south bands that stretched across the forest patch perpendicular to the prevailing winds that came from the ocean side of the patch, and four additional collectors were placed randomly within the first 30 m of the windward edge. In the fog season of 2004 (June-October), we sampled throughfall weekly from each of the 44 throughfall collectors. In the rain season, October 2004 through April 2005, we sampled throughfall biweekly from two randomly selected collectors per band to characterize each band. Two additional open (bulk) rain collectors were placed outside the forest south-west of the forest stand, and these were sampled year-round. Fog water was collected outside the forest using a passive fog collector with a plastic mesh collection surface (after Azevedo and Morgan 1974). For each sampling period, water was combined in the field by band for chemical analysis. In the rain season, when water volumes were large, collections were volume-weighted.

We installed mixed ion exchange resin columns (following Simkin *et al*. 2004; Templer & McCann 2010; Templer & Weathers 2011) to measure rates of N inputs and $\delta^{15}N$ and $\delta^{18}O$ of NO_3^- (see below for details about isotope analysis) in the canopy and to the forest floor during the summer fog season between August and October 2006. We placed three resin columns in the canopy (minimum 10 m above-ground) of three individual redwood trees (n = 9 resin columns in the canopy total; hereafter referred as ‘canopy collector’) and three resin columns at the base of the same three individual redwood trees (n = 9 resin columns on the ground total; hereafter referred to as ‘throughfall collector’). The 10 m height for our canopy collections was chosen so that we could obtain an integrative measure of atmospheric inputs throughout the canopy. One of the trees is at the edge of the forest stand, while the other two are in the interior. The three collectors within the canopy of each tree were located at 120^{O} horizontally from each other. Each resin column consisted of a 20-cm-diameter funnel attached to a 20-mL disposable chromatography column and set on top of a vertical PVC pipe. Tree climbers installed each canopy collector two meters from the bole of the tree by attaching the vertical PVC pipe to another two-metre-long horizontal pipe. Thus, the resin collectors in the canopy received throughfall from the canopy above without direct interference from the bole of the tree. Each collector on the forest floor had the same construction as those in the canopy, but the vertical pipe was placed directly in the ground as with the aqueous throughfall collectors. The 20-mL disposable chromatography columns were packed with Dowex Monosphere MR-3 UPW mixed ion exchange resin and had a 30-µm pore-size filter at the bottom of each column. Poly wool was placed at the neck of the funnel to prevent debris from entering the resin column. The funnel collected rainwater and canopy throughfall and channelled it through the resin column where charged nitrogen ions from the throughfall were adsorbed to the resin.

Nitrogen in soil solution

We also took advantage of samples collected in tension and gravimetric lysimeters from June 2004 through April 2005 and reported by Ewing *et al*. (2009) to measure natural abundance ^{15}N in NH_4^+ and NO_3^- in soil solution. Briefly, two tension lysimeters (Soil Moisture 1900 series) were installed at 12 cm depth (mineral soil horizon) 2 m apart in each of the five forest bands, and tension was set at 50 kPa after each sampling. Two zero-tension (gravimetric) lysimeters were installed at 70 cm depth (mineral soil horizon) in soil pits close to tension lysimeters in each of the five bands and near bulk collectors (see Ewing *et al*. 2009 for details). Tension and gravimetric lysimeters samples within each band were combined (keeping tension and gravimetric lysimeter samples separate) and collected on the same day as fog, bulk and throughfall collections. During the rainy season, volume-weighted 500 mL subsamples from gravimetric lysimeters were used since a larger volume was often found in these collectors.

We placed six ion exchange resin bags in the soil at a depth of 15 cm to measure nitrogen in soil solution at the base of the trees used to measure canopy inputs during the summer fog season between August and October 2006 (n = 2 resin bags paired with each throughfall collector; n = 18 resin bags total). Bags were placed in the soil by inserting a flat bar at an angle to create a slit extending into the mineral soil. The resin bags were placed in this slit to a depth of 15 cm, which was below the zone of high fine root density. Each resin bag was packed with 10 g of Dowex Monosphere MR-3 UPW mixed ion exchange resin. The amount of inorganic nitrogen adsorbed to the resin was used as a proxy for the mobility of the inorganic nitrogen ions in soil solution (Giblin *et al*. 1994).

Laboratory analyses

Water samples were analysed for nitrogen concentrations and reported in Ewing *et al*. (2009). All water samples were filtered (glass A/E filter) within 1 day of collection and refrigerated at 4 °C or frozen until analysis. Water samples were prepared for $\delta^{15}N$ isotope analysis by diffusion (Herman *et al*. 1995). We used KNO_3 and $(NH_4)_2SO_4$ (range of 20–150 µg N) as standards that were diffused with samples. N-isotope ratios were measured using an automated ANCA SL elemental analyser (Europa Scientific Ltd., Cheshire, UK) coupled with a Thermo Delta Plus XL mass spectrometer (Thermo Fisher Scientific, Waltham, MA, USA). The isotope ratio is expressed in ‘delta’ notation (‰), where the isotopic composition of any measured material relative to that of a standard on a per mil deviation basis is given by $\delta^{15}N = (R_{sample}/R_{standard}\text{-}1) \times 1000$, where R is the molecular ratio of heavy-to-light isotope forms. The standard for carbon is V-PDB. The standard for nitrogen is air. The reference material NIST SMR 1547, peach leaves, was used as calibration standard. These isotope analyses were conducted at the Center for Stable Isotope Biogeochemistry at the University of California, Berkeley.

Extraction of resin columns and bags was done within 24 h of collecting samples from the field. For resin extraction, we added 50 mL of 2M KCl to the 10 g resin beads three times sequentially (a total of 150 mL) and filtered with Whatman #1 filter paper. NH_4^+, NO_3^- and NO_2^- (NO_3^- and NO_2^- hereafter referred to as NO_3^-) concentrations in filtrate were determined on a Lachat QuikChem 8000 flow injection analyser (Lachat-Zellweger Instruments, Milwaukee, WI, USA). We used the salicylate method (E10-107-0602-A) for NH_4^+ concentrations in solution and the NO_3^- 8000 method (E10-107-04-1-C) for determining NO_3^- concentrations in solution. We used an external calibration standard, Environmental Resource Associates catalogue #505, in addition to an internal standard curve. Atmospheric nitrogen inputs were converted to fluxes using the concentration of the extractant solution, the total volume of extractant (150 mL KCl), the dimensions of the funnel (20 cm diameter) attached to each resin collector and total number of days incubated in the field (64 days). To quantify nitrogen in soil solution, we converted nitrogen concentrations to nitrogen accumulation rates (mg N g resin^{-1} day^{-1}) using the total volume of extractant (150 mL KCl), total mass of resin bag per bag (10 g) and the total number of days bags were incubated in the field (64 days). See below for details of stable isotope analysis of ^{15}N and ^{18}O of NO_3^- in resin extract samples.

Partitioning sources of nitrate in lysimeters and resin bags (soil solution)

We estimated the relative contributions of atmospheric inputs and nitrification in soil solution from our resin bag and lysimeter samples using the natural abundance isotope composition of NO_3^- in a two-end-member mixing model (Pardo *et al*. 2004; Templer & McCann 2010). ^{15}N values alone cannot be used to partition sources of NO_3^- between atmospheric inputs and biological processes such as nitrification because the ^{15}N abundance of atmospherically derived and microbially produced $^{15}NO_3^-$ overlap (Kendall & McDonnel 1998). In contrast to $\delta^{15}N$, $\delta^{18}O$ of NO_3^- differs significantly between NO_3^- of precipitation and NO_3^- produced microbially during nitrification. Therefore, the ^{18}O signature of NO_3^- in throughfall, soil solution or leachate can be used in a two-end-member mixing model to determine the predominant source of NO_3^- moving through the canopy to the forest floor in a terrestrial ecosystem (Böttcher *et al*. 1990; Durka *et al*. 1994; Kendall *et al*. 1996; Willard *et al*. 2001; Burns & Kendall 2002; Campbell *et al*. 2002; Ohte *et al*. 2004; Pardo *et al*. 2004; Templer *et al*. 2015).

We measured natural abundance ^{15}N and ^{18}O of nitrate in water samples collected in the summer fog season of 2005, as well as resin samples collected during the fog season of 2006 (see above for details of sampling). Throughfall water samples from the forest interior were bulked across four interior bands and were analysed separately from forest edge samples. The denitrifier method (Casciotti *et al*. 2002; Templer & Weathers 2011) was used to determine the natural abundance isotope composition of NO_3^- in

water and resin samples for fog water, canopy water, throughfall and soil solution of those samples that had sufficient NO_3^- concentrations (> 0.1 mg N L^{-1}). The stable isotope composition of nitrogen and oxygen in nitrous oxide gas produced by the bacteria was measured on a SerCon Cryoprep trace gas concentration system interfaced to a PDZ Europa 20–20 isotope ratio mass spectrometer (SerCon Ltd., Cheshire, UK) at the University of California, Davis Stable Isotope Facility. We used USGS standards #32, 34 and 35. Long-term precision for this analysis is ± 0.015 ‰ for $\delta^{15}N$ and 0.0093 ‰ for $\delta^{18}O$ ‰.

The following equation was used to partition the sources of NO_3^- in the soil solution between atmospheric inputs and nitrification:

$$\%NO_3^- \text{ from atmospheric inputs} = \left(\frac{\delta^{18}O_{loss} - \delta^{18}O_{nitrification}}{\delta^{18}O_{input} - \delta^{18}O_{nitrification}}\right) * 100 \quad (1)$$

where $\delta^{18}O_{loss}$ is the value of $\delta^{18}O$ in soil solution, $\delta^{18}O_{input}$ is the value of $\delta^{18}O$ in atmospheric inputs, and $\delta^{18}O_{nitrification}$ is the published value of $\delta^{18}O$ for nitrification (Kendall & McDonnel 1998; Pardo *et al*. 2004).

Natural abundance isotopes of redwood leaves and roots

Foliage was collected in August 2004 at a height of 45 m from four locations facing west (windward), east, north and south on one tree at the forest edge and one tree in the forest interior (180 m from forest edge; $n = 2$ trees total). Roots were collected on the same day from four sides of two trees at the windward edge and forest interior ($n = 4$ trees total). Foliage and root samples were dried at 65 ˚C, homogenized with a mortar and pestle, and analysed for carbon and nitrogen concentrations, and $\delta^{15}N$ and $\delta^{13}C$ using an automated ANCA SL nitrogen–carbon analyser coupled with an Europa 20/20 isotope ratio mass spectrometer (Europa Scientific, Crewe, UK) at the Center for Stable Isotope Biogeochemistry at the University of California, Berkeley. The reference material NIST SMR 1547, peach leaves, was used as calibration standard.

Natural abundance $\delta^{13}C$ of leaf sugars was determined on leaves made by the tree in 2003 and 2004 following the methods outlined in Hynson *et al*. (2012). The leaves were sampled from three forest edge and three forest interior redwood trees. The leaves used for the sugar $\delta^{13}C$ analyses were growing in full sun within 2 m of the tree tops. After the leaves were collected in the field between 15:00 and 16:00, they were frozen and transported back to the laboratory where soluble sugars were extracted for carbon isotope ratio analysis. One mg of samples was combusted in an elemental analyser interfaced with a Finnigan MAT Delta plus XL isotope ratio mass spectrometer (Thermo Instruments, Bremen, Germany) at the Center for Stable Isotope Biogeochemistry, University of California, Berkeley. Long-term precision for this analysis is ± 0.12 ‰.

Photosynthesis and uptake of Nitrogen by redwoods

Following the procedures outlined and tested in Ambrose, Sillett & Dawson (2009), leaf carbon assimilation (photosynthesis) rates were determined using a LiCor 6400 portable gas exchange system (Lincoln, NE, USA) two times in the year – the rainy period (November–April) and summertime fog period (July–August) in 2003 and 2004. The leaves were sampled from the same three forest edge and forest interior redwood trees sampled for sugar $\delta^{13}C$ analyses. Three small branches, each pointing in a different compass direction and about 90–95° from the other, were obtained for three edge and three interior trees. A series of leaves (12–14 lying flat in parallel that were attached to leafy shoots) were inserted into the standard cuvette. These small (40–60 cm long) branches were cut from the upper tree crown (sun exposed; 2–4 m from the very apex of each tree) and immediately brought to the ground, recut under water and allowed to rehydrate for at least four hours in very low light before measurements were taken. The cuvette conditions were held constant at a temperature of 18–20 °C, 1300 μmol m^{-2} s^{-1} of light supplied by the LiCor RED-BLUE light source and a $[CO_2]$ of 375 ppm. After measurements were completed, the leaf area and mass of each set of measured leaves were determined.

Plant uptake of nitrogen by fine roots was measured in August 2004 (fog season) adjacent to three forest edge and three interior trees (n = 6 trees total) using the *in situ* N depletion method with intact roots of redwood trees (Rennenberg *et al*. 1996; Gessler *et al*. 1998). We sampled roots < 2 mm in diameter since fine roots are the most active in root nutrient acquisition (Eissenstat 1992). Roots were located in the Oa horizon within a 5 m radius of each target redwood tree. Terminal root branches (< 2 mm diameter) for each target tree were excavated at the soil surface, taking care to ensure that roots remained intact and connected to the tree. Roots were rinsed with de-ionized water and gently blotted with Kimwipes® to remove soil particles from the root surface. Following excavation, while remaining attached to the tree, each fine root sample was placed in its own centrifuge tube containing 1.5 mL of treatment solution containing known concentrations of NH_4^+ and NO_3^-. Roots were incubated for 60 min in one of eight nutrient solutions: 0, 25, 50, 75, 100, 150, 200 and 250 μmol NH_4^+ and NO_3^- (i.e. 1:1 molar ratio of N in NH_4^+ and NO_3^-). The range of inorganic nitrogen in treatment solutions overlapped with nitrogen solution concentrations used in other studies of nitrogen uptake by trees (Lucash *et al*. 2007; Kahmen, Livesley & Arndt 2009; Socci & Templer 2011). Each solution tube was covered with parafilm during the 60-minute incubation to reduce evaporation and contamination. A set of tubes containing nutrient solutions, but no roots, was incubated as a control in each stand to determine evaporative losses and contamination of the solutions during the incubation period. At the end of the 60-minute incubation period, roots were removed from the solution and the incubated portion of the root was cut at the solution surface. Roots were oven-dried at 55 °C for 4 days and weighed. Treatment solutions were taken to the

laboratory where they were immediately filtered using Whatman #1 filters that were pre-washed with de-ionized water to remove any residual nitrogen contamination from manufacturing.

To determine concentrations of NH_4^+ and NO_3^- in each nutrient solution following the 60-minute incubation, we used a Lachat QuickChem 8000 Flow Injector Analyzer. To determine nutrient uptake rates, we calculated the change in nitrogen mass per tube by calculating the change in nutrient concentration between the beginning and end of the 60-min incubation period and the volume of solution. The rates of nitrogen uptake are expressed as net uptake, in which positive values represent a net uptake of nutrients by roots and negative values represent net efflux of nutrients from the roots.

We used the following equation to determine rates of NH_4-N and NO_3-N uptake:

$$N_{uptake} = [(N_{initial} * Vol_{solution}^{-1}) - (N_{final} * Vol_{solution}^{-1})] * g dw_{root}^{-1} * hour^{-1} \quad (2)$$

where N_{uptake} is the net uptake of N by fine roots over time; $N_{initial}$ = concentration of nitrogen in the treatment solution at the start of incubation; N_{final} = concentration of nitrogen in the treatment solution at the end of incubation; $Vol_{solution}$ = volume of the treatment solution; and g dw_{root} = dry root mass.

Statistical analyses

We conducted a one-way analysis of variance (ANOVA) (i) using forest location (i.e. edge vs. interior) as the main effect and nitrogen and carbon concentration, $\delta^{15}N$ and $\delta^{13}C$, and C:N ratios of redwood leaves or roots, as well as mass-based rates of photosynthesis as response variables, (ii) using nitrogen pool (i.e. bulk precipitation, summer fog, throughfall, gravimetric and tension lysimeters) as the main effect and natural abundance ^{15}N in NH_4^+ and NO_3^- as response variables within the fog or rain season, and (iii) using sample type (bulk precipitation, fog, throughfall, gravimetric and tension lysimeters for water samples and canopy, throughfall and soil solution for resin samples) as the main effect and $\delta^{15}N$ and ^{18}O in NO_3^- as the response variables. We conducted a two-way ANOVA using sample type (i.e. canopy vs. forest floor throughfall) and forest location (forest edge vs. interior) as the main effects and NH_4-N, NO_3-N or total inorganic N fluxes in 2006 as the response variables. We used Tukey's HSD test for *post hoc* comparisons of means when there was a statistically significant difference in the mean. We used SAS JMP PRO software (Version 11.2.0, 2013) for all statistical analyses.

Results

Within the fog season, inputs of NH_4^+, NO_3^- and total inorganic nitrogen in throughfall were greater at the forest edge compared to the forest interior ($P < 0.05$), but measurements in the canopy compared to the forest floor were

not significantly different from each other ($P > 0.05$; Fig. 1). NO_3^- and total inorganic N concentrations were also greater in soil solution at the forest edge compared to the forest interior ($P < 0.05$; Fig. 1).

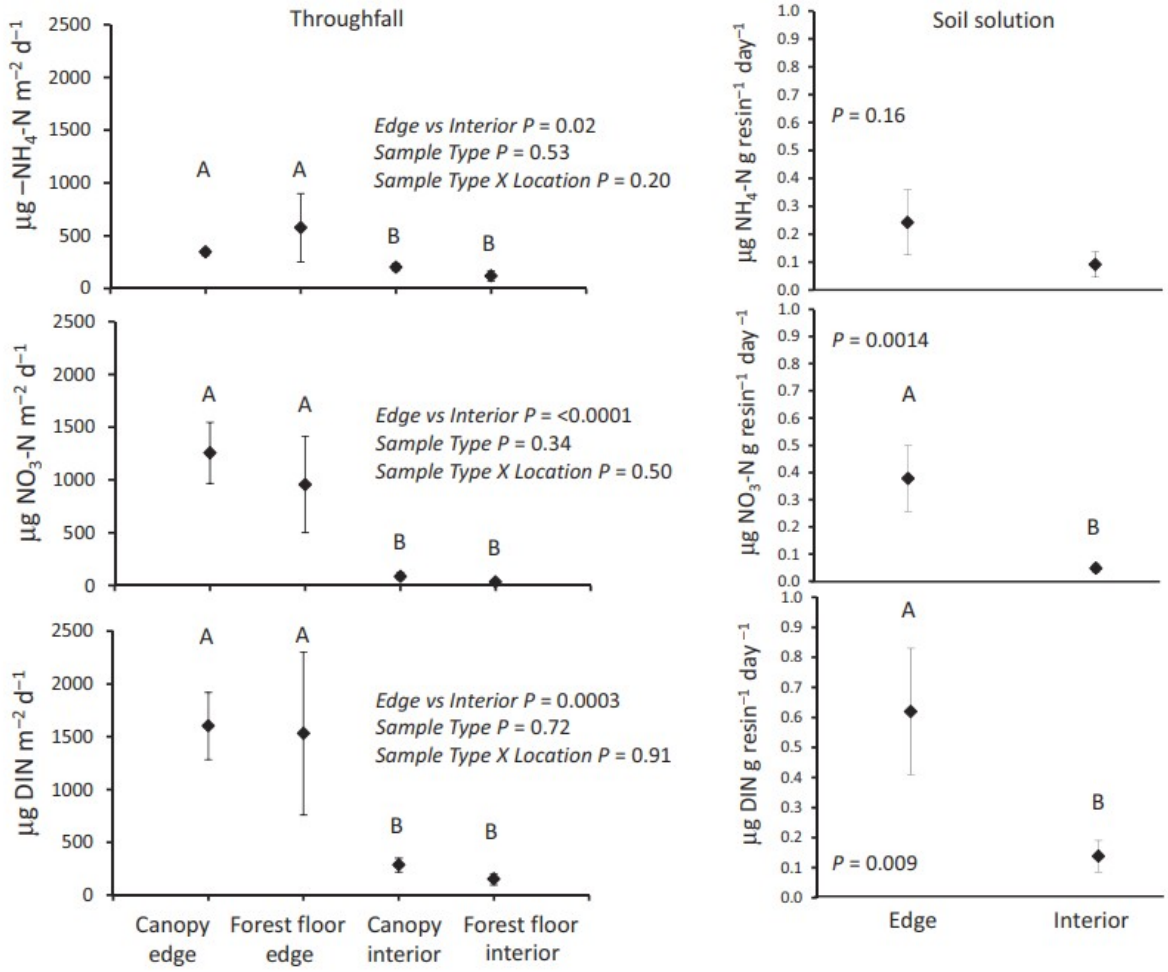

Fig. 1. Nitrogen inputs as throughfall measured in the canopy and forest floor, as well as soil solution during the fog season (measured in resin columns and bags) from a redwood forest in Sonoma, California. Values are means with standard error, and different letters indicate statistically significant ($P < 0.05$) differences in fluxes.

Natural abundance ^{15}N in NH_4^+ in water samples was enriched relative to NO_3^- across both the fog and rain seasons (Table 1). While $\delta^{15}NO_3$ N did not differ significantly among sample types within the fog or rain seasons, $\delta^{15}NH_4$ in fog was significantly greater than that in throughfall in the fog season. Isotopic values for aqueous samples between edge and interior locations did not differ significantly.

Table 1. Natural abundance ^{15}N in NH_4^+ and NO_3^- in water samples collected in the fog and rain seasons from a redwood forest in Sonoma, California. Values are means with standard error. Different letters indicate statistically significant differences in $\delta^{15}NH_4^+$ within a given season. Isotopic values for throughfall include all locations within the forest since there was no statistically significant difference in $\delta^{15}NH_4^+$ or $\delta^{15}NO_3^-$ between the forest edge and interior within the summer fog or winter rain seasons ($P > 0.05$). Missing values are due to insufficient volume of water to measure $\delta^{15}NH_4^+$ or $\delta^{15}NO_3^-$

	$\delta^{15}NH_4^+$ (‰)		$\delta^{15}NO_3^-$ (‰)	
Sample type	Fog season	Rain season	Fog season	Rain season
Fog	11.36[a] ± 2.68	8.72[a] ± 2.00	−2.63 ± 0.97	−0.91 ± 0.80
Throughfall	3.94[b] ± 0.98	2.60[a] ± 0.92	−2.76 ± 0.89	−0.95 ± 0.65
Bulk		1.21[a] ± 0.88		−0.54 ± 0.69
Gravimetric lysimeter		16.70[b] ± 0.74		−0.71 ± 0.81
Tension lysimeter			−1.52 ± 3.61	−0.65 ± 1.40
P value	0.02	<0.0001	0.56	1

To determine the proportion of NO_3^- coming directly from precipitation (i.e. bulk precipitation in winter and fog in summer) vs. nitrification in the canopy or forest floor, we used a two-end-member mixing model (equation 1 above) for both water and resin samples. For water sample end members, we used the mean value calculated for $\delta^{18}O$ in bulk precipitation in winter (66.8 ‰; Fig. 2a) or fog in summer (68.1 ‰) as atmospheric inputs and the mean value for $\delta^{18}O$ in gravimetric and tension lysimeters (1.7 ‰) as soil solution. For resin sample end members, we used the mean value calculated for $\delta^{18}O$ in canopy collectors (76.6 ‰) as atmospheric inputs and resin bags (32.4 ‰) for soil solution. For both water and resin samples, we used the published range of $\delta^{18}O$ values for NO_3^- produced from nitrification (−5 to +15 ‰; Kendall *et al*. 1996; Pardo *et al*. 2004). Within the lysimeter samples, between 91 and 100% NO_3^- in soil solution comes from nitrification rather than directly from atmospheric inputs in both summer and winter. Within our resin bag samples, between 54 and 66% NO_3^- in soil solution comes from microbial production in soils rather than directly from atmospheric inputs in summer.

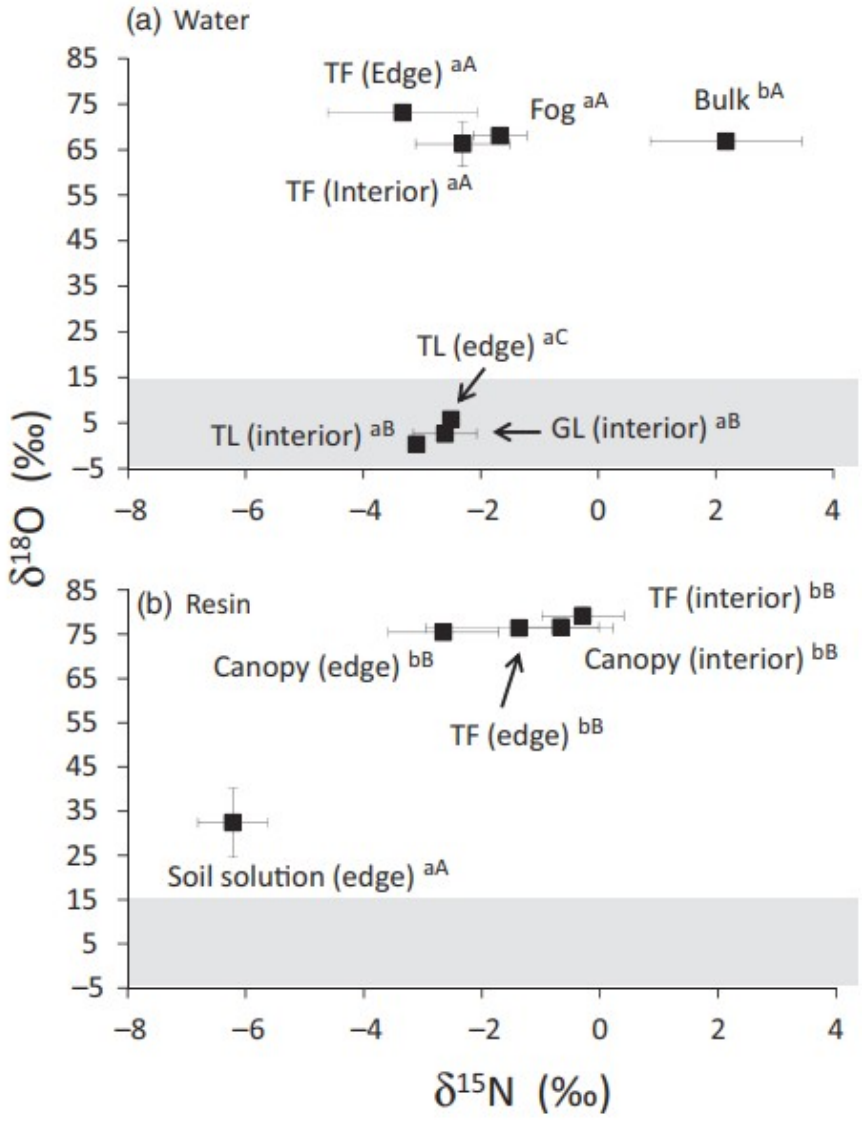

Fig. 2. Natural abundance ^{15}N and ^{18}O in (a) water samples (fog water, bulk precipitation, throughfall, tension lysimeters and gravimetric lysimeters) and (b) resin column and resin bag (soil solution) samples collected in the summer fog season from a redwood forest, Sonoma, California. Values are means with standard error. TF = throughfall; TL = tension lysimeter; GL = gravimetric lysimeter. Different lower and uppercase letters within each figure indicate statistically significant differences in $\delta^{15}N$ ($^0/_{00}$) or $\delta^{18}O$ ($^0/_{00}$), respectively. Grey-shaded area indicates published range of $\delta^{18}O$ values for NO_3^- produced from nitrification (−5 to +15 $^0/_{00}$; Kendall *et al.* 1996; Pardo *et al.* 2004). There was not a sufficient amount of NO_3^- in resin bag soil solution samples from the forest interior for ^{15}N and ^{18}O analysis.

Isotopic signatures of NO_3^- sampled in the summer fog season using ion exchange resin varied across sample types. Natural abundance ^{18}O in NO_3^- was greater in water samples from atmospheric inputs (i.e. throughfall, fog and bulk precipitation) compared to those in the soil solution samples we obtained from gravimetric and tension lysimeters (Fig. 2a). Similarly, $\delta^{18}O$ was greater in the canopy and throughfall resin samples compared to soil solution samples (Fig. 2b). Natural abundance ^{15}N was significantly greater in bulk precipitation compared to throughfall, fog and lysimeters water samples (Fig. 2a). However, bulk precipitation in summer represents < 1% of annual precipitation rates in this forest (Ewing *et al*. 2009). Finally, $\delta^{15}N$ was significantly greater in canopy and throughfall resin column extracts compared to soil solution water samples collected in the resin bags (Fig. 2b).

Nitrogen and carbon concentrations, as well as $\delta^{15}N$ and $\delta^{13}C$, were all higher in the leaves of edge trees compared to forest interior trees ($P < 0.0001$), but we found no significant difference in N and C concentrations, C:N ratios or $\delta^{15}N$ and $\delta^{13}C$ in the roots of edge vs. forest interior trees (Table 2). The C:N ratios of forest edge tree leaves were significantly lower than those of forest interior trees ($P < 0.0001$). Natural abundance $\delta^{13}C$ of leaf sugars and rates of photosynthesis were not significantly different between the forest edge and interior during the rain season ($P > 0.05$; Fig. 3). In contrast, leaf sugar $\delta^{13}C$ ($P = 0.022$) and rates of photosynthesis ($P = 0.001$) were both statistically significantly higher at the forest edge compared to the interior in the fog seasons in both years examined. Rates of photosynthesis were significantly lower in interior trees during the fog season compared to both edge and interior trees during the rain season of both years ($P = 0.016$). The edge trees in the fog season had significantly greater $\delta^{13}C$ in leaf sugars than both the edge and interior trees in the rain season in both years ($P = 0.003$). There were no significant differences in NH_4^+ vs. NO_3^- uptake with forest location, though there was a trend towards greater nitrogen uptake by redwood roots at the forest edge compared to the interior of the forest for NH_4^+ ($P = 0.10$), NO_3^- ($P = 0.27$) and total inorganic N ($P = 0.096$; Fig. 4).

Table 2. Nitrogen, carbon and isotopic abundance of leaves and roots collected. Values are means with standard error

Location	%N	$\delta^{15}N$	%C	$\delta^{13}C$	C:N
Leaves (edge)	1.17 ± 0.05	−1.27 ± 0.37	51.28 ± 0.04	−27.37 ± 0.08	44.37 ± 1.64
Leaves (interior)	0.76 ± 0.01	−4.09 ± 0.13	49.20 ± 0.23	−28.79 ± 0.21	64.86 ± 1.03
P value	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
Roots (edge)	1.14 ± 0.05	−2.39 ± 0.25	48.83 ± 0.16	−26.84 ± 0.44	43.12 ± 2.10
Roots (interior)	1.26 ± 0.02	−2.92 ± 0.33	49.50 ± 0.34	−27.24 ± 0.08	39.46 ± 0.36
P value	0.085	0.24	0.13	0.40	0.14

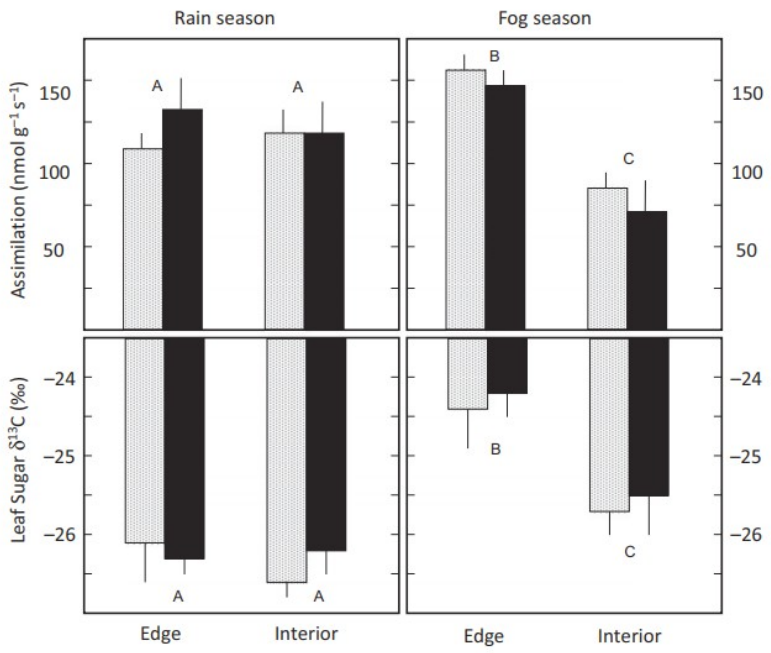

Fig. 3. Leaf carbon assimilation rate (photosynthesis) expressed on a leaf mass basis (upper panel) and natural abundance stable carbon isotopes ($\delta^{13}C$) of leaf sugars. Leaves sampled from redwood trees at forest edge (n = 3 trees) and interior (n = 3 trees) during the rain and fog season in 2003 (grey bars) and 2004 (black bars; lower panel) from a redwood forest, Sonoma, California. Bars are means with standard error. Different letters within upper and lower panel indicate statistically significant differences in rates of photosynthesis and leaf sugar $\delta^{13}C$, respectively.

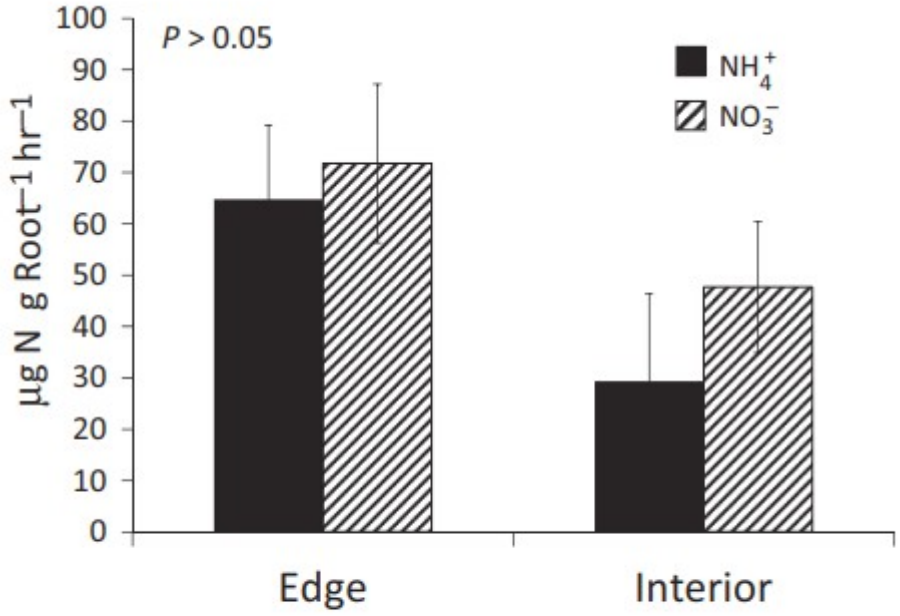

Fig. 4. Root nitrogen uptake of NH_4^+ and NO_3^- by redwood trees at the forest edge and interior (180 m from forest edge) from a redwood forest, Sonoma, California. Values are means with standard error.

Discussion

Summer fog as a source of nitrogen for redwood trees: the forest edge vs. the interior

Multiple lines of evidence from this study show that summer fog contains nitrogen that is available for redwood tree uptake and that such uptake probably occurs through their foliage (Figs 1 and 5). First, similar to past studies (Ewing *et al*. 2009), we found greater fluxes of throughfall nitrogen to the forest floor and in the soil solution at the west-facing, most fog-inundated forest edge compared to the interior, suggesting that trees at the windward side of this forest experience greater nitrogen availability. Secondly, in agreement with our expectations, we found that leaves within the edge of the coastal redwood forest had greater concentrations of nitrogen and higher $\delta^{15}N$ compared to foliage in the interior of the forest stand. Since $^{15}NH_4^+$ values in fog are significantly enriched compared to bulk precipitation and throughfall inputs, it appears that fog is a likely source of nitrogen. These patterns are not surprising as the potential for horizontal interception of fog within forests is greater at the forest edge than the interior (Weathers, Lovett & Likens 1995; 2001, Ewing *et al*. 2009; del Val *et al*. 2006). In contrast, we did not find significant differences in root $\delta^{15}N$ or rates of NH_4^+ and NO_3^- uptake by roots between the forest edge and interior, suggesting that patterns of greater foliar $\delta^{15}N$ at the forest edge compared to the interior reflect direct uptake of nitrogen in fog by redwood leaves. While it is not possible to discern the relative proportion of nitrogen taken up via foliage vs. roots, our results suggest that at least in a location with substantial fog input such as this coastal redwood forest, natural abundance ^{15}N in leaves is influenced by foliar uptake of fog-borne nitrogen.

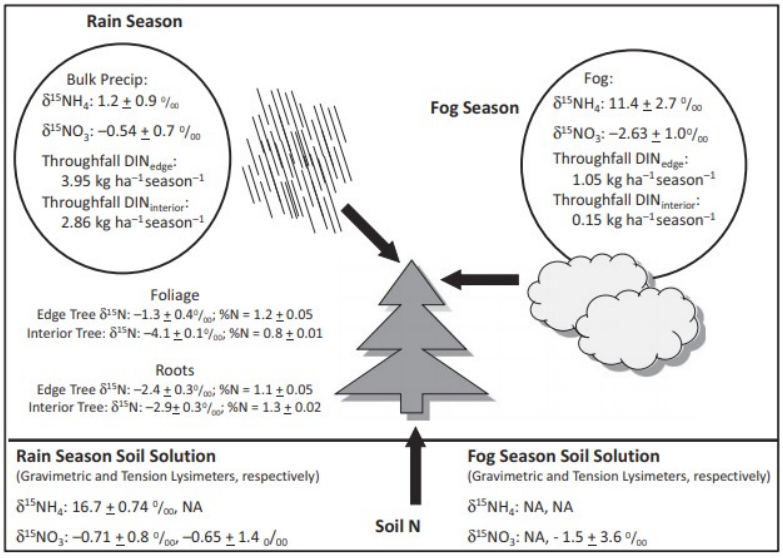

Fig. 5. Conceptual model for plant nitrogen uptake and natural abundance ^{15}N in water samples and plant tissue of a coastal redwood forest, based on a redwood forest, Sonoma County, California. The diagram shows horizontal flow of fog during summer months and vertical flow of rain in winter. Nitrogen fluxes for throughfall from Ewing *et al.* (2009). NH_4^+ and NO_3^- make up approximately one-third and two-thirds, respectively, of total inorganic nitrogen fluxes to the forest floor when averaged across seasons and forest location. NH_4^+ and NO_3^- make up approximately 18% and 82%, respectively, of fog. NA indicates insufficient water volume for $\delta^{15}N$ analysis.

The greater nitrogen concentrations of foliage at the forest edge (Table 2) could be due to greater nitrogen inputs from fog or could be explained in part by possibly greater rates of mineralization and nitrification in soils, which were not measured in this study. The lower C:N ratios found in foliage at the forest edge may translate into lower C:N and more labile litterfall,

leading to greater rates of mineralization and nitrification compared to the forest interior. Given that tension lysimeters in the upper portion of the soil profile collected more nitrogen at the edge than interior site, though total nitrogen flux to depths > 70 cm was greater at the interior site (Ewing *et al*. 2009), it seems likely that trees at the forest edge have access to a greater amount of nitrogen from the soil compared to forest interior trees. However, we did not find significant differences in rates of nitrogen uptake by roots at the forest edge compared to the interior.

Fog water also contributes to shifts in the physiological activity of trees. The higher rates of photosynthesis and more enriched $\delta^{13}C$ of leaf soluble sugars at the forest edge compared to the interior during the fog season (Fig. 3) could be attributed to the edge trees having greater rates of uptake of water (Dawson 1998) and greater foliar nitrogen invested in enzymes, leading to improved water-use efficiency (Dawson *et al*. 2002) compared to trees in the forest interior. Past work by Ewing *et al*. (2009) shows that trees at the edge of this forest also have greater canopy wetness, total leaf area and crown water uptake (as evidenced by reverse sap flow).

It is possible that the benefits of summer fog are limited to the edge of coastal redwood forest ecosystems, but our and others' data from the forest interior suggest that fog water and the nutrients it contains have strong effects on ecosystem structure and function and these influences are not limited to the forest edge. Ewing *et al*. (2009) showed that the half-deposition distance of nitrogen from fog in this redwood forest is approximately 30 m into the forest, meaning the distance that throughfall nitrogen fluxes are half the rate of the windward edge, which is similar to edge effect half-deposition distances found in other studies (Weathers, Lovett & Likens 1995). While there is a strong edge effect of fog on below canopy processes such as throughfall fluxes of nitrogen, the canopy of the forest interior probably benefits from fog water and nitrogen throughout the summer months. Evidence for decoupling between canopy processes and throughfall water inputs to the forest floor is provided by sustained canopy wetness and foliar water uptake in the forest interior throughout the summer fog season (Ewing *et al*. 2009). It is also possible that nitrogen uptake from fog during the summer months (whether by roots or the canopy itself) is elevated throughout the forest from edge to interior compared to what it would be without any nitrogen inputs from fog.

Atmospheric or biological source of nitrate in the canopy and forest floor

The presence of organic soil within the canopy of some large redwood trees (Enloe, Graham & Sillett 2006) could contribute to biogeochemical cycling of nitrogen within the canopy of these forest ecosystems, therefore providing an additional source of nitrogen for redwood trees. Similar to throughfall to the forest floor, throughfall inputs of NH_4^+ and NO_3^- within the forest canopy itself are also larger at the forest edge than the interior (Fig. 1). However, the lack of difference in $\delta^{18}O$ between throughfall and fog or bulk water

samples, as well as between canopy and throughfall resin samples (Fig. 2), indicates that there was little canopy production of NO_3^- via nitrification. Recent work shows very low rates of microbial decomposition of organic matter in soil mats of redwood canopies, which has been attributed to low pH, low moisture and poor litter quality (Enloe *et al*. 2010). Our results suggest that NO_3^- produced (if any) within these arboreal mats is either not produced in large quantities or is immobilized by epiphytic plants or microbes before it enters the forest floor as throughfall. Thus, the relatively large flux of throughfall NO_3^- within the forest edge compared to the interior (Fig. 1) and the large pool of NO_3^- in the redwood canopy observed in past studies (Ewing *et al*. 2009) probably comes directly from atmospheric sources, rather than from microbial production within the canopy.

In contrast to the canopy, the significantly depleted $\delta^{18}O$ values in soil solution (lysimeters and resin bags) compared to atmospheric canopy inputs (i.e. fog, bulk precipitation and throughfall) indicate nitrification by soil microbes, rather than direct atmospheric inputs from fog or bulk precipitation, as a primary source of NO_3^- in soils within both the rain and fog season. The greater proportion of NO_3^- coming directly from canopy inputs (rather than from nitrification) in the resin bags compared to the lysimeters was surprising, but may be explained by both the length of sampling and the difference in soil depth at which soil solution is measured. The resin bags were left in the field to collect soil solution for six weeks and provided an integrative measure over time, while the lysimeters samples provided a snapshot of nitrogen in soil solution. Also, it is possible that placement of resin bags directly adjacent to redwood trees could have funnelled fog-derived nitrogen down to the resin bags more effectively than to the lysimeters, which were placed randomly with respect to the location of redwood trees.

Conclusions

The idea that fog is important to coastal redwood forest distribution and productivity has long been asserted (Noss 2000; Mooney & Dawson 2015), but the mechanisms and details of how and where in forest ecosystems fog affects ecosystem functions have been more elusive. Results of this study show that the nitrogen carried within fog is important and can be utilized by redwood trees, especially those at the forest edge. Coastal redwood trees are able to take advantage of greater nitrogen and water availability at the forest edge, leading to greater rates of photosynthesis in summer. Further, the isotopic evidence presented here shows that the substantial NO_3^- inputs to the forest floor from the canopy as throughfall, especially at the forest edge, comes directly from atmospheric inputs (i.e. fog or rain) rather than in-canopy nitrification.

While the relatively small sample sizes prevent broad generalizations to the entire range of coastal redwood forests throughout the western United

States, this study provides the most direct evidence to date of redwood canopy utilization of fog nitrogen inputs. Although we found greater variability in throughfall fluxes using ion exchange resin collectors compared to water sampling, we detected significant differences across the forest edge compared to the interior, suggesting that our sample size was sufficient to discern the effects of forest location on throughfall nitrogen inputs both within the canopy and to the forest floor. Similarly, we only sampled leaves for natural abundance ^{13}C and measured photosynthesis from three forest edge and three forest interior trees, and these were sufficient sample sizes to detect differences in photosynthetic activity and products among forest locations. It is possible that patterns we observed could be due to differences between forest edge and interior that were independent of fog inputs, but the lack of difference in water inputs, rates of photosynthesis or $\delta^{13}C$ in sugars in the rain season suggests that it is more likely that fog inputs in summer are important in causing the differentiation between edge and interior sites.

Future research is needed to determine how much nitrogen is taken up by roots vs. directly into leaves or small twigs of redwood trees and how these relate to soil nitrogen cycling rates at the forest edge and interior. Together, these efforts would enable researchers to partition soil solution vs. direct fog uptake of nitrogen and the contribution soil turnover of nitrogen and fog inputs make to nitrogen uptake by redwood trees.

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Data accessibility

Data for Tables 1 and 2, as well as Figs 1-4, are available on DRYAD, Provisional DOI: doi:10.5061/dryad.0j2q8.

References

Ambrose, A.R., Sillett, S.C. & Dawson, T.E. (2009) Effects of tree height on branch hydraulics, leaf structure, and gas exchange in California's redwood species. *Plant, Cell and Environment*, 32, 743– 757.

Azevedo, J. & Morgan, D.L. (1974) Fog precipitation in coastal California forests. *Ecology*, 55, 135– 1141.

Barbosa, O., Marquet, P.A., Bacigalupe, L.D., Christie, D.A., del-Val, E., Gutierrez, A.G., Jones, C.G., Weathers, K.C. & Armesto, J.J. (2010) Interactions among patch area, forest structure and water fluxes in a fog-inundated forest ecosystem in semi-arid Chile. *Functional Ecology* 24, 909– 917.

Beyn, F., Matthias, V. & Dahnke, K. (2014) Changes in atmospheric nitrate deposition in Germany – an isotopic perspective. *Environmental Pollution*, 194, 1– 10.

Böttcher, J., Strebel, O., Voerkelius, S. & Schmidt, H.L. (1990) Using isotopic fractionation of nitrate-nitrogen and nitrate-oxygen for the evaluation of microbial denitrification in a sandy aquifer. *Journal of Hydrology*, 114, 413–424.

Bowden, R.D., Geballe, G.T. & Bowden, W.B. (1989) Foliar uptake of ^{15}N from simulated cloud water by red spruce (*Picea rubens*) seedlings. *Canadian Journal of Forest Research*, 139, 383– 386.

Bradbury, D.C. & Firestone, M.K. (2007) Environmental control of microbial N transformations in redwood forests. In: (eds R.B. Standiford, G.A. Giusti, Y. Valachovic, W.J. Zielinski & M.J. Furniss), pp. 203– 204. *Proceedings of the Redwood Region Forest Science Symposium: What Does the Future Hold?* Gen. Tech. Rep. PSW-GTR-194. Pacific Southwest Research Station, Forest Service, U.S. Department of Agriculture, Albany, CA, USA.

Burgess, S.S.O. & Dawson, T.E. (2004) The contribution of fog to the water relations of *Sequoia sempervirens* (D Don): foliar uptake and prevention of dehydration. *Plant, Cell and Environment*, 27, 1023– 1034.

Burns, D.A. & Kendall, C. (2002) Analysis of sources of $\delta^{15}N$ and $\delta^{18}O$ to differentiate NO_3^- sources in runoff at two watersheds in the Catskill Mountains of New York. *Water Resources Research*, 38, 9-1– 9-11.

Campbell, D.H., Kendall, C., Chang, C.C.C.Y., Silva, S.R. & Tonnessen, K.A. (2002) Pathways for nitrate release from an alpine watershed: determination using $\delta^{18}O$ and $\delta^{15}N$. *Water Resources Research*, 38, 10– 1029.

Casciotti, K.L., Sigman, D.M., Hastings, M.G., Ohlke, J.K. & Hilkert, A. (2002) Measurement of the oxygen isotopic composition of nitrate in seawater and freshwater using the denitrifier method. *Analytical Chemistry*, 74, 4905–4912.

Clark, K., Nadkarni, N.M., Schaefer, D. & Gholz, H.L. (1998) Atmospheric deposition and net retention of ions by the canopy in a tropical montane forest, Monteverde, Costa Rica. *Journal of Tropical Ecology*, 14, 27– 45.

Dawson, T.E. (1998) Fog in the California redwood forest: ecosystem inputs and use by plants. *Oecologia*, 117, 476– 485.

Dawson, T.E., Mambelli, S., Plamboeck, A.H., Templer, P.H. & Tu, K.P. (2002) Stable isotopes in plant ecology. *Annual Review of Ecology and Systematics*, 33, 507– 559.

Durka, W., Schulze, E., Gebauer, G. & Voerkelius, S. (1994) Effects of forest decline on uptake and leaching of deposited nitrate determined from ^{15}N and ^{18}O measurements. *Nature*, 372, 765– 767.

Eissenstat, D.M. (1992) Costs and benefits of constructing roots of small diameter. *Journal of Plant Nutrition*, 15, 763– 782.

Elliott, E.M., Kendall, C., Wankel, S.D., Burns, D.A., Boyer, E.W., Harlin, K., Bain, D.J. & Butler, T.J. (2007) Nitrogen isotopes as indicators of NO_x source contributions to atmospheric nitrate deposition across the Midwestern and northeastern United States. *Environmental Science and Technology*, 41, 7661– 7667.

Enloe, H.A., Graham, R.C. & Sillett, S.C. (2006) Arboreal Histosols in old-growth redwood forest canopies, northern California. *Soil Science Society of America Journal*, 70, 408– 418.

Enloe, H.A., Quideau, S.A., Graham, R.C., Sillett, S.C., Oh, S.W. & Wasylishen, R.E. (2010) Soil organic matter processes in old-growth redwood forest canopies. *Soil Science Society of America Journal*, 74, 161– 171.

Evans, R.D. & Ehleringer, J.R. (1994) Plant $\delta^{15}N$ values along a fog gradient in the Atacama Desert, Chile. *Journal of Arid Environments*, 28, 189– 193.

Ewing, H.A., Weathers, K.C., Templer, P.H., Dawson, T.E., Firestone, M.K., Elliott, A.M. & Boukili, V.K.S. (2009) Fog water and ecosystem function: heterogeneity in a California redwood forest. *Ecosystems*, 12, 417– 433.

Felix, J.D., Elliott, E.M., Gish, T.J., McConnell, L.L. & Shaw, S.L. (2013) Characterizing the isotopic composition of atmospheric ammonia emission sources using passive samplers and a combined oxidation-bacterial denitrifier approach. *Rapid Communications in Mass Spectrometry*, 27, 2239– 2246.

Fenn, M.E., Poth, M.A., Schilling, S.L. & Grainger, D.B. (2000) Throughfall and fog deposition of nitrogen and sulfur at an N-limited and N-saturated site in the San Bernardino Mountains, southern California. *Canadian Journal Forest Research*, 30, 1476– 1488.

Gessler, A., Schneider, S., Von Sengbusch, D., Weber, P., Hanemann, U., Huber, C., Rothe, A., Kreutzer, K. & Rennenberg, H. (1998) Field and laboratory experiments on net uptake of nitrate and ammonium by the roots of spruce (*Picea abies*) and beech (*Fagus sylvatica*) trees. *New Phytologist*, 138, 275– 285.

Giblin, A.E., Laundre, J.A., Nadelhoffer, K.J. & Shaver, G.R. (1994) Measuring nutrient availability in Arctic soils using ion-exchange resins: a field test. *Soil Science Society of America*, 58, 1154– 1162.

González, A.L., Fariña, J.M., Pinto, R., Pérez, C., Weathers, K.C., Armesto, J.J. & Marquet, P.A. (2011) Bromeliad growth and stoichiometry: responses to

atmospheric nutrient supply in fog-dependent ecosystems of the hyper-arid Atacama Desert, Chile. *Oecologia*, 167, 835– 845.

Harrison, A.F., Schulze, E.D., Gebauer, G. & Bruckner, G. (2000) Canopy uptake and utilization of atmospheric pollutant nitrogen. *Carbon and Nitrogen Cycling in European Forest Ecosystems Ecological Studies*, 142, 171– 188.

Hastings, M.D., Casciotti, K.L. & Elliott, E.M. (2013) Stable isotopes as tracers of anthropogenic nitrogen sources, deposition and impacts. *Elements*, 9, 339– 344.

Heaton, T.H.E. (1990) Nitrogen-15/nitrogen-14 ratios of NO_x from vehicle engines and coal-fired power stations. *Tellus, Series B: Chemical and Physical Meteorology*, 42B, 304– 307.

Herman, D.J., Brooks, P.D., Ashraf, M., Azam, F. & Mulvaney, R.L. (1995) Evaluation of methods for nitrogen-15 analysis of inorganic nitrogen in soil extracts. II. Diffusion methods. *Communications in Soil Science and Plant Analysis*, 26, 1675– 1685.

Hiatt, C., Fernandez, D. & Potter, C. (2012) Measurements of fog water deposition on the California Central Coast. *Atmospheric and Climate Sciences*, 2, 525– 531.

Hietz, P., Wolfgan, W., Wania, R. & Nadkarni, N. (2002) Nitrogen-15 natural abundance in a montane cloud forest canopy as an indicator of nitrogen cycling and epiphyte nutrition. *Oecologia*, 131, 350– 355.

Hynson, N.A., Mambelli, S., Amend, A.S. & Dawson, T.E. (2012) Shady business: stable isotopes, and severing mycorrhizal networks show changes in mycoheterotrophy under light limitation. *Oecologia*, 169, 307– 317.

Igawa, M., Tsutsumi, Y., Mori, T. & Okochi, H. (1998) Fogwater chemistry at a mountainside forest and the estimation of the air pollutant deposition via fog droplets based on the atmospheric quality at the mountain base. *Environmental Science and Technology*, 32, 1566– 1572.

Kahmen, A., Livesley, S.J. & Arndt, S.K. (2009) High potential, but low actual, glycine uptake of dominant plant species in three Australian land-use types with intermediate N availability. *Plant and Soil*, 325, 109– 121.

C. Kendall & J.J. McDonnel (eds.) (1998) *Isotope Tracers in Catchment Hydrology*. Elsevier Science B.V, Amsterdam, The Netherlands.

Kendall, C., Silva, S.R., Chang, C.C.Y., Burns, D.A., Campbell, D.H. & Shanley, J.B. (1996) Use of the d18O and d15N of nitrate to determine sources of nitrate in early spring runoff in forest catchments. *Symposium on Isotopes in Water Resources Management, Vienna, Austria*. 20–24 March 1995. IAEA-SM-336/29. International Atomic Energy Agency, Vienna, Austria, pp. 167– 176.

Kimball, K.D., Jagels, R., Gordon, G.A., Weathers, K.C. & Carlisle, J. (1988) Differences between New England coastal fog and mountain cloud water chemistry. *Water, Air, and Soil Pollution*, 39, 383– 393.

Lucash, M.S., Eissenstat, D.M., Devereux Joslin, J., McFarlane, K.J. & Yanai, R.D. (2007) Estimating nutrient uptake by mature tree roots under field conditions: challenges and opportunities. *Trees*, 21, 593– 603.

Mooney, H.A. & Dawson, T.E. (2015) Coast redwood forests of California. *Ecosystems of California* (eds E. Zavaleta & H.A. Mooney), Chapter 26. University of California Press, Berkeley, CA, USA.

Moore, H. (1977) The isotopic composition of ammonia, nitrogen dioxide and nitrate in the atmosphere. *Atmospheric Environment*, 11, 1239– 1243.

Noss, R., ed. 2000. *The Redwood Forest: History, Ecology, and Conservation of the Coast Redwoods*. Island Press, Washington, DC, USA.

Ohte, N., Sebestyen, S.D., Shanley, J.B., Doctor, D.H., Kendall, C., Wankel, S.D. & Boyer, E.W. (2004) Tracing sources of nitrate in snowmelt runoff using a high-resolution isotopic technique. *Geophysical Research Letters*, 31, L21506.

Pardo, L.H., Kendall, C., Pett-Ridge, J. & Chang, C.C.Y. (2004) Evaluating the source of stream water nitrate using $\delta^{15}N$ and $\delta^{18}O$ in nitrate in two watersheds in New Hampshire, USA. *Hydrological Processes*, 18, 2699– 2712.

Petterssen, S. (1936) On the causes and the forecasting of the California fog. *Journal of Aeronautical Sciences*, 3, 305– 309.

Rennenberg, H., Schneider, S. & Weber, P. (1996) Analysis of uptake and allocation of nitrogen and sulfur compounds by trees in the field. *Journal of Experimental Botany*, 47, 1491– 1498.

Santiago, L.S. & Dawson, T.E. (2014) Light use efficiency of California redwood forest understory plants along a moisture gradient. *Oecologia*, 174, 351– 363.

Simkin, S.M., Lewis, D.N., Weathers, K.C., Lovett, G.M. & Schwarz, K. (2004) Determination of sulfate, nitrate and chloride in throughfall using ion-exchange resins. *Water, Air and Soil Pollution*, 153, 343– 354.

Socci, A.M. & Templer, P.H. (2011) Temporal patterns of inorganic nitrogen uptake by mature sugar maple (*Acer saccharum* Marsh.) and red spruce (*Picea rubens* Sarg.) trees using two common approaches. *Plant Ecology and Diversity*, 4, 141– 152.

Sparks, J.P. (2009) Ecological ramifications of the direct foliar uptake of nitrogen. *Oecologia*, 159, 1– 13.

Templer, P.H., Ewing, H., Weathers, K., Dawson, T. & Firestone, M. (2006) *Fog as a Potential Source of Nitrogen for Coastal Redwood Forest*

Ecosystems. American Geophysical Union annual meeting. Moscone Center, San Francisco, CA, USA. December 11–15, 2006.

Templer, P.H. & McCann, T.M. (2010) Effects of the Hemlock Woolly Adelgid on Nitrogen Losses from Urban and Rural Northern Forest Ecosystems. *Ecosystems*, 13, 1215– 1226.

Templer, P.H. & Weathers, K.C. (2011) Use of mixed ion exchange resin and the denitrifier method to determine isotopic values of nitrate in atmospheric deposition and canopy throughfall. *Atmospheric Environment*, 45, 2017–2020.

Templer, P.H., Weathers, K.C., Lindsey, A., Lenoir, K. & Scott, L. (2015) Atmospheric inputs and nitrogen saturation status in and adjacent to Class I wilderness areas of the northeastern United States. *Oecologia*, 177, 5– 15.

United States Department of Agriculture (USDA) Forest Service and Soil Conservation Service. (1972) Soil survey Sonoma county California.

del Val, E., Armesto, J.J., Barbosa, O., Christie, D.A., Gutiérrez, A.G., Jones, C.G., Marquet, P.A. & Weathers, K.C. (2006) Rain forest islands in the Chilean semiarid region: fog-dependency, ecosystem persistence and tree regeneration. *Ecosystems*, 9, 598– 608.

Weathers, K.C. & Likens, G.E. (1997) Clouds in Southern Chile: an important source of nitrogen to nitrogen-limited ecosystems. *Environ Science and Technology*, 31, 210– 213.

Weathers, K.C., Lovett, G.M. & Likens, G.E. (1995) Cloud deposition to a spruce forest edge. *Atmospheric Environment*, 29, 665– 672.

Weathers, K.C., Likens, G.E., Bormann, F.H., Bicknell, S.H., Bormann, B.T., Daube, B.C. Jr *et al*. (1988) Cloudwater chemistry from ten sites in North America. *Environmental Science and Technology*, 22, 1018– 1026.

Weathers, K.C., Lovett, G.M., Likens, G.E. & Caraco, N.F.M. (2000) Cloudwater inputs of nitrogen to forest ecosystems in southern Chile, forms, fluxes, and sources. *Ecosystems*, 3, 590– 595.

Willard, K.W.J., DeWalle, D.R., Edwards, P.J. & Sharpe, W.E. (2001) ^{18}O isotopic separation of stream nitrate sources in mid-Appalachian forested watersheds. *Journal of Hydrology*, 252, 174– 188.