Aerosol and Air Quality Research, 18: 1953–1992, 2018

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2018.03.0107

Review

Impacts and Effects Indicators of Atmospheric Deposition of Major Pollutants to Various Ecosystems - A Review

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ABSTRACT

In this paper, we review the current understanding on ecosystem and human health impacts from the atmospheric deposition of acidifying pollutants, eutrophying nitrogen (N), polycyclic aromatic hydrocarbons (PAHs), mercury (Hg), trace metals, and ozone (O_3), as well as the biological indicators that have been used to assess the health of ecosystems following exposure to these pollutants. We provide overviews of the impacts of deposition for these pollutants and discuss the currently known biomonitors for each pollutant. The deposition of acidifying pollutants impacts terrestrial ecosystems by altering plant physiology and growth and by increasing plant susceptibility to stresses that can be indirectly damaging to the health of fish and birds. Indicators of the deposition of acidifying pollutants include soil base cation content and acid neutralizing capacity, among others. Eutrophying N deposition has been studied extensively; N enrichment directly impacts vegetative plant species cover, richness, growth rates, and susceptibility to other stressors. It indirectly impacts wildlife through changes in their habitats and food sources. Indicators for N deposition include changes in plant species and in tissue and litter N content. The deposition of PAHs has been found to cause significant damage to plant organisms and to be carcinogenic and mutagenic to humans and animals. Useful biomonitors of PAH deposition include lichens, mosses, and pine needles. Deposited Hg can undergo methylation (in the presence of sulphur reducing bacteria); bioaccumulation of methylmercury is highly toxic to animals. Effective biomonitors of Hg contamination of aquatic ecosystems are fish and marine birds. The impacts of O_3 are well understood, with well-established "flux" models being vast improvements on the previous AOT40 approaches. This review highlights the impacts that the above-mentioned pollutants have on terrestrial and aquatic organisms and the biomonitors that are currently being used to assess the deposition levels and effects of these pollutants.

Keywords: Air pollution; Atmospheric deposition; Ecosystem health; Ecological monitoring.

INTRODUCTION

Atmospheric deposition is a major source of pollutants to terrestrial (e.g., forests, grasslands, ombrotrophic wetlands) and aquatic (rivers, lakes, oceans) environments (Lepori and Keck, 2012). Other non-depositional processes (e.g., effluent discharge, run-off, leaching) are also important pathways for pollutant input, especially to aquatic ecosystems (Vodyanitskii, 2013). Atmospheric deposition of pollutants can cause detrimental effects to both terrestrial and aquatic environments and to human and wildlife health (Driscoll *et al.*, 2001; Baldigo *et al.*, 2009; Bobbink *et al.*, 2010; Bacon *et al.*, 2013; Vodyanitskii, 2013; de Vries *et al.*, 2014; Jones *et al.*, 2014; de Vries *et al.*, 2015; Duan *et al.*, 2016; Rodríguez-Estival and Smits, 2016). Once deposited onto land and water surfaces, pollutants are incorporated into soil or water where they can be taken up directly by biota, methylated by microorganisms in the case of mercury (Hg), or increase the leaching of soil base cations, metals, and plant nutrients in the case of acidifying pollutants. Moreover, pollutants can also be transported from where they were initially deposited via soil erosion, surface run-off, and groundwater flows. Physical, chemical and biological variables can affect pollutant uptake and toxicity in biota. Exposure to pollutants can lead to wide-ranging ecological consequences. Methylmercury is a neurotoxin capable of bio-accumulating in wildlife and biomagnifying in food

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webs; humans are typically exposed via fish or other wildlife consumption (Wiener *et al.*, 2012). Depletion of plant nutrients and leaching of toxic inorganic aluminum due to acid rain can damage vegetation, increase stress to and reduce tolerance of plants, and inhibit tree growth. Acidified lakes are also detrimental to aquatic wildlife (Driscoll *et al.*, 2001). Polycyclic aromatic compounds (PACs) and its subgroup of polycyclic aromatic hydrocarbons (PAHs) comprise hundreds of hydrocarbons, some of which are cancer-causing agents or capable of causing damage to DNA or reproductive impairment (Kim *et al.*, 2013; Abdel-Shafy and Mansour, 2016).

Despite the knowledge that deposition occurs, monitoring of emissions, ambient air, and deposition is often conducted separately from the monitoring of ecosystem effects leading to gaps in the understanding of sources and fate of pollutants in the environment (Ochoa-Hueso *et al.*, 2017). In the former case, researchers are able to track the variability in the emissions, air concentrations and deposition but are uncertain about the post-deposition effects and how these changes will impact ecosystems. In the latter, researchers are able to assess the ecosystem effects, but are uncertain what the sources of contamination are and the role of atmospheric deposition versus the physical, chemical, and biological factors influencing pollutant uptake. These knowledge gaps may be filled by establishing stronger connections between atmospheric sources or deposition and ecological effects. This can be accomplished through the identification of biomonitors and their associated effects from pollutant exposure and in the determination of the critical loads and critical levels at which these effects occur. A biomonitor is defined as a change in biological responses, ranging from molecular through cellular and physiological responses to behavioral changes, which can be related to exposure to, or toxic effects of, environmental chemicals (Peakall and Walker, 1994). A biological indicator refers to a living organism that provides information on an ecosystem. A critical load refers to a concentration limit in any media (air, water, soil, tissue). In this review paper, we examine the ecosystem and human health impacts from atmospheric deposition of acidifying pollutants, eutrophying nitrogen (N), polycyclic aromatic hydrocarbons (PAH), Hg, trace metals, and ozone (O_3), as well as the biological indicators that have been used to assess the health of ecosystems. Deposition studies on other groups of pollutants have also been made in literature (Lin *et al.*, 2010; Weissengruber *et al.*, 2018), but the information was not as rich as for those pollutants mentioned above, and thus were not included in this review.

ACIDIC DEPOSITION

Major Acidic Species

Major acidic species in the atmosphere include gases (SO_2, HNO_3, HCl, and organic acids) and particulate matters (sulphate, nitrate, chloride, organic acids), and associated aqueous-phase species (Seinfeld and Pandis, 2016). The components of acidic deposition vary based on the local and regional emissions but primarily consist of sulphur species. The relative contribution of each pollutant to total acidic deposition also depends on meteorology (e.g., amount of precipitation, sunlight, wind speed), which can impact formation rates as well as deposition velocities. Typically, oxidized sulphur (i.e., SO_2 and SO_4^{2-}) and nitrogen (primarily HNO_3 and NO_3^-) dominate acidic deposition in polluted areas (Vet *et al.*, 2014). The deposition of reduced nitrogen ($NH_x = NH_3 + NH_4^+$) can also acidify terrestrial ecosystems (soils) through two indirect mechanisms: (i) NH_4^+ uptake into roots (which releases H^+), or (ii) nitrification to NO_3^- (which produces H^+) (Krupa, 2003). Base cation species (e.g., Ca^{2+}, Mg^{2+}, Na^+, K^+) can mitigate acidic deposition by neutralizing acidic species, and should be taken into account when assessing acidification of receiving ecosystems (Watmough *et al.*, 2014). The characteristics of acidifying pollutants and base cations, as well as commonly used indicators, are summarized in Table 1.

Impacts of Acidic Deposition

It is well established that acidic (sulphur and nitrogen) deposition can decrease soil pH, and increase soil solution concentrations of Al^{3+}, resulting in the leaching of base cations (Ca^{2+}, Mg^{2+}, and K^+). Direct effects from deposition on leaves are manifested in changes in leaf physiology, including an increase in leaf roundness (Bacon *et al.*, 2013), changes in the epicuticular and epistomatal waxes (Bartiromo *et al.*, 2012; Elliott-Kingston *et al.*, 2014), smaller leaves, and changes to the cuticle and stomata (Haworth *et al.*, 2010; Haworth *et al.*, 2012; Elliott-Kingston *et al.*, 2014). Impacts on forests include nutrient deficiencies due to soil base cation leaching (Akselsson *et al.*, 2007), leading to decreased growth (Savva and Berninger, 2010), changes in tree ring width (Godek *et al.*, 2015), increased susceptibility to disease, destruction of root systems resulting in tree and crown dieback (Wilmot *et al.*, 1995; Godek *et al.*, 2015), and higher sensitivity to drought (Savva and Berninger, 2010). The effects of acid deposition have been found to have a geographical dependence (de Vries *et al.*, 1995). Regional properties of an ecosystem that have been found to relate to acid deposition include soil chemistry, such as texture, pH, and humidity, and forest properties, such as tree species and age (de Vries *et al.*, 1995; Økland *et al.*, 2004; de Vries *et al.*, 2007; Stevens *et al.*, 2009a; van Dobben and de Vries, 2010).

Impacts on aquatic ecosystems from acidic deposition are fairly well understood; the mobilisation of Al from soils and decreased water pH can lead to reduced fish species richness, changes in fish health, and degraded water quality. The reduction and extinction of Atlantic salmon in the Maritimes (Watt *et al.*, 2000) and fish populations elsewhere (Monteith *et al.*, 2005; Gray *et al.*, 2012) have been associated with increased water acidity.

Acidic deposition may impact wildlife by reducing the microbial biomass and shredder (macroinvertebrates that feed on plant and animal material and break it into smaller particles) biomass of gammarids (Meegan *et al.*, 1996) and caddisflies (Simon *et al.*, 2009), suppressing microbial respiration (Dangles and Guérold, 2001; Simon *et al.*, 2009), depressing aquatic fungi (Clivot *et al.*, 2014), damaging the gills of fish with gill lesions due to increases in Al and

Table 1. Characteristics of Acidifying Pollutants (N & S) and Base Cations (Ca^{2+}, Mg^{2+}, K^+, Na^+).

Element	Acidifying Pollutants		Base Cations (Alkaline)			
	Nitrogen (N)	Sulfur (S)	Calcium (Ca^{2+})	Magnesium (Mg^{2+})	Potassium (K^+)	Sodium (Na^+)
Atomic Number	7	16	20	12	19	11
Group	15	16	2	2	1	1
Period	2	3	4	3	4	3
Atomic Mass	14.00067 g mol^{-1}	32.06 g mol^{-1}	40.08 g mol^{-1}	24.305 g mol^{-1}	39.0983 g mol^{-1}	22.98977 g mol^{-1}
Series	Nonmetal	Nonmetal	Alkali Earth Metal	Alkali Earth Metal	Alkali Metal	Alkali Metal
Interesting	Inert in elemental form	Occurs naturally near volcanoes	• Occurs only in compounds (because it is so reactive)			
Main Sources	On- and Off-Road vehicles, the oil and gas industry, and fuel for electricity generation and heating	Smelting and refining non-ferrous metals, coal-fired electricity generation, and the petroleum industry	• Dust associated with mining activities. • Minerals like chalk, limestone, and marble. • Atmospheric deposition and weathering. • Deposition fertilizer	• Deposition of fertilizer	• Weathering of primary materials • Deposition of fertilizer	
Common Compounds	• Organic Nitrogen Compounds (ONC) • Nitric Oxide (NO) • Nitrogen Oxides (NO_x) • Nitrogen Dioxide (NO_2) • Nitric Acid (HNO_3) • Particulate Nitrate (NO_3^-) • Particulate Nitrite (NO_2^-) • Ammonia (NH_3) • Ammonium (NH_4^+)	• Sulfuric Acid (H_2SO_4) • Sulphur Oxides (SO_x) • Sulphur Dioxide (SO_2) • Sulphate(SO_4^{2-})	• Limestone ($CaCO_3$) • Gypsum ($CaSO_4 \cdot 2H_2O$) • Fluorite (CaF_2) • Calcium Chloride ($CaCl_2$)	• Magnesium Chloride ($MgCl_2$)	• Potassium Chloride (KCl)	• Sodium Chloride (NaCl) • Sodium Nitrate ($NaNO_3$)
Indicators	• N concentration in plants • Tissue N content • Species richness • Forb richness • Graminoid:Forb ratio • Shifts in plant communities • Shifts in diatom communities • Nitrate concentrations • C:N ratio in soil • Dissolved nitrogen • Dissolved phosphorus	• S content • S:N ratio • Tree leaves • Tree rings • Physiognomic leaf analysis • Soil percent base saturation • Acid neutralizing capacity	• Base cation surplus in soil • Ca:Al^{3+} ratios in soil • (Ca+Mg):S ratio in throughfall • Foliar chemistry • Soil water chemistry • pH • Inorganic Al^{3+} • Calcium			

decreases in calcium (Evans *et al.*, 1988), and thinning of bird egg shells (Hames *et al.*, 2002). Effects such as those mentioned above can both directly and indirectly affect macroinvertebrates (Baldigo *et al.*, 2009; Ferreira and Guérold, 2017). Tables 2, 3, and 4 summarize the key points of these studies that examine the impacts of N, S, and soil base cation deposition, respectively.

Effects Indicators of Acidic Deposition

Effects indicators of acidification on terrestrial ecosystems include S and N tissue concentrations (Wieder *et al.*, 2016); tree ring widths (Lee *et al.*, 2011; Godek *et al.*, 2015; van der Sleen *et al.*, 2015); tree bark acidity (Grodzińska, 1977); and the molar ratio of Ca:Al^{3+} (or base cation (BC):Al^{3+}) in soil solution (de Vries, 1993; Vanguelova *et al.*, 2007; Kleijn *et al.*, 2008; Jorge *et al.*, 2011; de Vries *et al.*, 2015; Irvine *et al.*, 2017), where a ratio < 1 implies a high risk and > 10 implies low risk; soil pH (Kleijn *et al.*, 2008; Tian and Niu, 2015); soil solution Al^{3+}, exchangeable base cation (BC) content in soil (Lucas *et al.*, 2011; Tian and Niu, 2015); soil percent base saturation, with increased risk at < 10% and lower risk at > 30%; foliar chemistry, with limited sugar maple growth at levels of Ca < 5000 ppm and Mg < 700 ppm (Fenn *et al.*, 2011); leaf physiogenomic traits (Bacon *et al.*, 2013); leaf litter content (Meegan *et al.*, 1996; Simon *et al.*, 2009; Clivot *et al.*, 2014); cuticle micromorphology (Bartiromo *et al.*, 2012); and isotopic ratio fingerprinting (Hogan *et al.*, 2017).

Indicators for aquatic ecosystems include acid neutralizing capacity (ANC = Σ base cations – Σ strong acid anions) where values lower than a threshold of 20 $\mu eq\ L^{-1}$ result in established risks to aquatic life (Lien *et al.*, 1996; Bulger *et al.*, 2000; Fölster *et al.*, 2007; Futter *et al.*, 2014; Duan *et al.*, 2016; Sullivan *et al.*, 2017); pH, with a reduction in fish species below a pH of 5.0–6.0 (Jeffries and Lam, 1993; Rask *et al.*, 1995; Futter *et al.*, 2014); inorganic Al^{3+}, with concentrations above 2 $\mu mol\ L^{-1}$ is deemed toxic to aquatic organisms (Sjostedt *et al.*, 2012); calcium, where concentrations below 1.5 mg L^{-1} negatively affect crustaceans (Wærvågen *et al.*, 2002); leaf-associated fungi (Clivot *et al.*, 2014); base cation surplus in streams (Baldigo *et al.*, 2009); and macroinvertebrates diversity using the acid biological assessment profile (acidPAPD) index (Baldigo *et al.*, 2009). In acidified streams, leaf litter analysis is a useful bioassessment tool on stream health as leaf litter decomposition rates increase with increasing acidity (Dangles and Guérold, 2001; Simon *et al.*, 2009; Ferreira and Guérold, 2017).

NITROGEN DEPOSITION

Major N-deposition Species

Similar to acidic deposition, the major components of N-deposition vary from region to region and depend on local emissions and meteorology. Generally, N-deposition is dominated by inorganic reduced nitrogen (NH_3 and NH_4^+) and oxidized nitrogen (NO_x, HNO_3 and NO_3^-) (e.g., Harrison *et al.*, 1999; Hayden *et al.*, 2003; Horii *et al.*, 2005; Zhang *et al.*, 2009; Benedict *et al.*, 2013; van den Elzen *et al.*, 2018). Although organic N deposition is not typically measured, it has been shown to constitute a significant fraction (upwards of ~30%) at some field sites (Zhang *et al.*, 2009; Benedict *et al.*, 2013). Due to the variety of atmospheric N species, as well as technical challenges measuring certain N species (e.g., NH_3 (von Bobrutzki *et al.*, 2010); organic N (Beem *et al.*, 2010); HONO (VandenBoer *et al.*, 2013)), complete accounting of total N deposition for a given ecosystem can be challenging.

Impacts of Nitrogen Deposition

Exposure to N can cause a wide variety of well documented impacts on terrestrial and aquatic ecosystems. Below the critical level, N can be beneficial to an environment, such as causing an increase in biomass and CO_2 uptakes (Butterbach-Bahl and Dannenmann, 2011; Jones *et al.*, 2014; Carter *et al.*, 2017; Xu *et al.*, 2017). However, above the critical level, the effects of N tend to be negative. Impacts include direct toxicity from dry deposition to individual species (Marschner *et al.*, 1996; Carter *et al.*, 2017), eutrophication, acidification (as discussed above), and negative impacts on plant species diversity owing to changes in the form of available N, such as oxidized N (from e.g., vehicle emissions, industry, and domestic combustion) and reduced N (from e.g., agricultural sources, biomass burning) (Southon *et al.*, 2013; Field *et al.*, 2014; Jones *et al.*, 2014; van den Berg *et al.*, 2016). Long-term exposure to terrestrial ecosystems can cause significant declines in plant cover including lichens (Bobbink *et al.*, 2010; Stevens *et al.*, 2012; Field *et al.*, 2014; Bähring *et al.*, 2017; Carter *et al.*, 2017) and mosses and herbaceous plants (Dirkse and Martakis, 1992; Hallingbäck and Kellner, 1992; Mountford *et al.*, 1993; Morecroft *et al.*, 1994; Mountford *et al.*, 1994; Kirkham *et al.*, 1996; Carroll *et al.*, 2000; Strengbom *et al.*, 2001; Pitcairn *et al.*, 2006; Koranda *et al.*, 2007; Bobbink *et al.*, 2010; Caporn *et al.*, 2014; Field *et al.*, 2014; Bähring *et al.*, 2017). However, N addition has also been found to cause an increase in the cover of N tolerant (nitrophilic) plant species (Verhoeven *et al.*, 2011; Field *et al.*, 2014; Bähring *et al.*, 2017). Impacts of N enrichment to grasslands include loss of species richness (Stevens *et al.*, 2004; Stevens *et al.*, 2006; Shepherd *et al.*, 2009; Stevens *et al.*, 2009b; Bobbink *et al.*, 2010; Sheppard *et al.*, 2011; Stevens *et al.*, 2012; Isbell *et al.*, 2013; Henry and Aherne, 2014; Bähring *et al.*, 2017), an increase in growth rate (Power *et al.*, 1995; Power *et al.*, 2006), an increase in flower formation (Power *et al.*, 1995; Power *et al.*, 2006; Bähring *et al.*, 2017), a decrease in soil C:N ratio (Bähring *et al.*, 2017) and an increase in shoot increments (Bähring *et al.*, 2017). Other implications include increased susceptibility to secondary stresses and disturbances, such as pests, drought, frost, and disease, and indirectly to exotic earthworm activity (Vitousek *et al.*, 1997; Bobbink *et al.*, 1998; Gilliam, 2006; de Vries *et al.*, 2015; Simkin *et al.*, 2016; Soons *et al.*, 2016; Shao *et al.*, 2017).

In forest ecosystems, impacts include changes in plant species diversity and decomposition inhibition; increases in canopy size, water shortages, tree mortality, soil acidity, and Al mobility; and decreases in Ca availability, fine-root biomass, understory plant cover, and dwarf-shrub species

Table 2. Impacts of Acidifying Pollutant: Nitrogen (N).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Terrestrial Ecosystems						
Calcareous dunes	Newborough Warren, North Wales, UK	Chronosequences and soil sampling Acidification	• Accumulation of soil C and N • Suppression of N_2 fixation • Leaching of excess N (low retention capacity for N in dune soils)	Chronosequences and soil sampling	Low N deposition. Regression modelling	Aggenbach *et al.*, 2016
Calcareous dunes	Luchterduinen, the Netherlands	Chronosequences and soil sampling Acidification	• Accelerated acidification • Decalcification of the topsoil	Chronosequences and soil sampling	High N deposition. Regression modelling	Aggenbach *et al.*, 2016
Acidic dunes	Luchterduinen, the Netherlands	Chronosequences and soil sampling Acidification	• Accelerated acidification. • Accelerated N accumulation rates	Chronosequences and soil sampling	High N deposition. Regression modelling	Aggenbach *et al.*, 2016
Calluna vulgaris (Dwarf shrubs)	Fehmarn Island, Baltic Sea, North Germany	6 levels of N fertilization	• Decrease of cover • Increase of shoot increments for low-N treatment • Increase in flower formation for high-N treatment	• Shoot increment most sensitive indicator • Soil C:N ratios deemed inappropriate indicator	Current critical N loads confirmed in study (3 yr study)	Bähring *et al.*, 2017
Carex arenaria and *Festuca ovina* (Graminoids)	Fehmarn Island, Baltic Sea, North Germany	6 levels of N fertilization	• Increase of cover • Decrease in C:N ratio		Current critical N loads confirmed in study (3 yr study)	Bähring *et al.*, 2017
Bryophytes	Fehmarn Island, Baltic Sea, North Germany	6 levels of N fertilization	• Decrease of cover • Decrease in C:N ratio		Current critical N loads confirmed in study (3 yr study)	Bähring *et al.*, 2017
Lichens	Fehmarn Island, Baltic Sea, North Germany	6 levels of N fertilization	• Decrease of cover • Decrease in C:N ratio for low-N treatment		Current critical N loads confirmed in study (3 yr study)	Bähring *et al.*, 2017
Sand dunes			• Sand dune binding by taller, more vigorous vegetation • Promotes higher dune building • Improves flood defences			Barbier *et al.*, 2008
Elymus Canadensis	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• No or poor emergence in field plots • Enhanced seed germination in greenhouse			Bird and Choi, 2016
Echinacea purpurea	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• No emergence in field plots • Enhanced seed germination in greenhouse			Bird and Choi, 2016

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Dalea purpurea	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• Reduced seed germination in greenhouse			Bird and Choi, 2016
Elymus	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• Reduced seed germination in greenhouse and no emergence in field plots.			Bird and Choi, 2016
Panicum virgatum	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• Increased germination in greenhouse • Increased germination in field plots in early years			Bird and Choi, 2016
Lupinus perennis	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• Low germination in greenhouse • Substantial establishment in field plots			Bird and Choi, 2016
Monarda punctata	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• Low germination in greenhouse • Substantial establishment in field plots			Bird and Choi, 2016
Schizachyrium scoparium	Lake Michigan, USA	N-addition to soil (15 and135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• Low germination in greenhouse • Substantial establishment in field plots			Bird and Choi, 2016
Rudbeckia hirta	Lake Michigan, USA	N-addition to soil (15 and 135 kg N ha^{-1} yr^{-1} NH_4NO_3)	• No emergence in field plots			Bird and Choi, 2016
Peat bogs	Europe	Peat litter decomposition	• Reduction in N limitations on microbial metabolism • Higher dissolved organic carbon release	• N:P ratio in litter • C:N ratio in litter		Bragazza *et al.*, 2006
Lichens and bryophytes	Europe (review previous studies)	Long-term NH_3 exposure			Annual critical level for lichens and bryophytes suggested to be 1 µg NH_3 m^{-3} for long-term (several years)	Cape *et al.*, 2009
Herbaceous plants	Europe (review previous studies)	Long-term NH_3 exposure			Annual critical level for herbaceous species suggested to be 3 ± 1 µg NH_3 m^{-3} for long-term (several years)	Cape *et al.*, 2009

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Calcifugous grasslands	UK	Long-term experimental N addition	Reduction in bryophyte cover and in cover of individual species			Carroll *et al.*, 2000
Lichens		N uptake measured				Dahlman *et al.*, 2004
Holycomium splendens	Europe	N eutrophication	Decline in species			Dirkse and Martakis, 1992; Hallingbäck and Kellner, 1992; Strengbom *et al.*, 2001; Koranda *et al.*, 2007
Forest floor vascular plants	Europe		Decrease in oligotrophic species with increasing CL exceedance			Dirnböck *et al.*, 2014
Grasslands			Acidification of soil and metal availability			Falkengren-Grerup *et al.*, 1987
Calcareous dunes (acid grassland bog, upland heath, lowland heath, and sand dune)	Great Britain	N deposition Acidification and eutrophication	• Decline in plant species richness • Changed species composition • Decline in diversity of mosses, lichens, and forbs • Increase in graminoids cover	• Decline in plant species richness • Changed species composition • Soil pH levels	• 135 locations surveyed (2 m × 2 m quadrats) • Decline in forb diversity below pH of 3.8 • Decline in acid grassland richness below pH of 4.5	Field *et al.*, 2014
					Review paper	Fowler *et al.*, 2013
			Particulate Nitrate (NO_3^-) Leaching causes acidification of soil			Gundersen and Rasmussen, 1990
	127 sampling sites in China		Higher leaf N concentrations correspond to high atmospheric N deposition.		(2094 observations in literature 1980–2005). Higher than critical load.	Han *et al.*, 2005
Nardus stricta and *Deschampia flexuosa*		Testing if vascular plants are feasible bio-indicators of atmospheric N deposition				Hicks *et al.*, 2000

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Mobile and semi-fixed coastal dunes	England and Wales, UK	N deposition gradient	• Increase in height and cover of *Ammophila arenaria*	No common indicator common to all dune sites	Treshold around 15 kg N ha^{-1} yr^{-1} suggested	Jones *et al.*, 2004
Fixed dunes	England and Wales, UK	N deposition gradient	• Increase in biomass • Increase in the cover of certain species • Reduced N availability	• Above ground biomass • Soil C:N ratio		Jones *et al.*, 2004
Grasslands and heathland	Europe				Review paper includes management and N budgets for grasslands and heathland	Jones *et al.*, 2017
Grasslands			Acidification of soil and metal availability			Jönsson *et al.*, 2003
Hippophae rhamnoides		N-fixing experiments	N_2 fixation can be switched off when mineral N in soil is sufficient.			Kato *et al.*, 2007
Temperate grasslands		Asymbiotic N_2 fixation				Keuter *et al.*, 2014
Calcareous Grey dunes	NW Europe		• Decline in plant species richness. Decalcification of top soil. • Grass encroachment not a major problem (1990–2014). Calcareous dunes less sensitive than acidic dunes. N-accumulation in soil unaffected by high N-deposition. Aeolian activity can bring calcareous sand back to the surface.		Extra N from ammonia emission from beaches. Critical load of 15 kg ha^{-1} yr^{-1} reached.	Kooijman *et al.*, 2016
Acidic Grey dunes	NW Europe		• Decline in plant species richness (functioning and species richness are very sensitive to high N-deposition). Grass encroachment. Net N-mineralization increased with N-deposition. • Grazing is needed to counteract grass encroachment	.	Critical load of 10 kg ha^{-1} yr^{-1} exceeded considerably due to extra ammonium being deposited from the sea.	Kooijman *et al.*, 2016

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Woody plant leaves	China		Higher leaf N concentrations correspond to high atmospheric N deposition.		981 observations in unfertilized and non-agricultural ecosystems	Liu *et al.*, 2013
Trifolium repens		N-fixing experiments	N_2 fixation can be switched off when mineral N in soil is sufficient.			Macduff *et al.*, 1996
Calcifugous grasslands	UK	Long-term experimental N addition	Decline in mosses after 3 years			Morecroft *et al.*, 1994
Unimproved hay meadow	Tandham Moor, Somerset	Low N addition (25 kg N ha^{-1} yr^{-1})	Significant decline in percent cover of *P. lanceolata*			Mountford *et al.*, 1993; Mountford *et al.*, 1994; Kirkham *et al.*, 1996
National Parks	Greater Yellowstone Area	Estimation of total N critical loads and development of atmospheric N deposition maps	Lowest CL and highest exceedances were located in high elevation basins with limited vegetation, exposed bedrock, and steep slopes		• Wet and dry deposition • CL for nutrient enrichment in aquatic ecosystems range from < 1.5 ± 1.0 kg N ha^{-1} yr^{-1} to > 4.0 ± 1.0 kg N ha^{-1} yr^{-1}	Nanus *et al.*, 2017
Forest (30 stands: sugar maple, red maple, American beech, yellow birch, paper birch, red spruce, balsam fir, eastern hemlock)	White Mountain, New Hampshire			• Foliar chemistry and soil N status provided positive feedback • Good soil C:N ratio prediction accuracy from canopy lignin:N ratio	Relationship observed between N-cycling rates and forest disturbance history	Ollinger *et al.*, 2002
Acidic iron-poor dunes	Danish coast		High grass encroachment.			Ovesen, 2001
Calluna vulgaris and *Erica cinerea*		Testing if vascular plants are feasible bio-indicators of atmospheric N deposition	Indicators of elevated N deposition and CL exceedance described and evaluated	Tissue N content		Pitcairn *et al.*, 1998; Pitcairn *et al.*, 2001; Pitcairn *et al.*, 2003
Ectohydric mosses	UK	N deposition	Loss of biodiversity	• Tissue N content of mosses excellent for sites where deposition is dominated by NH_3 • Tissue N content can be useful for identifying s	5 km × 5 km spatial resolution Spatial variability in upland areas related to cloud cover	Pitcairn *et al.*, 2006

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
				vegetation exposed to large concentrations of NH_4^+ or NO_3^- such as hilltops and wind exposed woodland edges	Tissue N appears to be more closely linked to rainfall NH_4^+ or NO_3^- concentrations than deposition.	
Calluna vulgaris		Testing if vascular plants are feasible bio-indicators of atmospheric N deposition			Urban-rural gradient	Power and Collins, 2010
Calluna vulgaris	UK	N addition (15.4 kg N ha^{-1} yr^{-1})	Increase in growth rates. Long-term increase in flower formation.		Lowland heathland	Power *et al.*, 1995; Power *et al.*, 2006
	UK	N deposition		• Mineral N leaching for high N deposition systems (midpoint indicator) • Tissue N concentration for low N deposition systems (midpoint indicator)	Metrics paper	Rowe *et al.*, 2016
Bog system (*Sphagnum capillifolium*, *Cladonia* spp. and *Calluna vulgaris*)		Ammonia release experiment	Loss of species richness – all disappeared at higher ammonia concentrations			Shepherd *et al.*, 2009
Calluna vulgaris, bryophytes, and sedge	UK	Gaseous NH_3 deposition	• Reduction and almost total loss in *Calluna vulgaris,Sphagnum capillifolium,*and *Cladonia portentosa* cover after 3 yrs • Increase in sedge cover		• Effects due to direct foliar uptake and interaction with abiotic and biotic stresses not effects on soil. • Effects on cover of sensitive species happen at lower NH_3 concentrations for longer exposure times	Sheppard *et al.*, 2011
Calluna vulgaris, bryophytes, and sedge	UK	Wet NH_4 deposition	• Significant increase in *Calluna vulgaris* cover after 5 yrs			Sheppard *et al.*, 2011

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
			• Reduction in *Sphagnum capillifolium*and *Cladonia portentosa* cover after 5 yrs			
Grasslands			Acidification of soil and metal availability			Skiba *et al.*, 1989
Herbaceous terrestrial and wetland ecosystems	China, Europe, the Americas	Long-term N enrichment experiments	Reduced plant species richness by an average 16%	• Presence or absence of indicator species found to not be useful as an indicator of N deposition impacts • Percentage cover of individual species suggested to have more promise as an indicator: *L.corniculatus,C. rotundifolia, H.splendens,P.lanceolata* and *T. tamariscinum*	189 field experiments, 51 sites. More pronounced reduction in China than Europe and the Americas	Soons *et al.*, 2016
Calcifugous Grasslands	England, Scotland, Wales (National gradient)		Acidification of soil and metal availability. Decline in forb and pleurocarpous moss richness.		Graminoid:forb ratio best indicator of N deposition impacts	Stevens *et al.*, 2006; Stevens *et al.*, 2009b
Lichen taxa	UK	N deposition	Declines in lichen taxa	Terricolous lichens as indicators	5 km × 5 km scale	Stevens *et al.*, 2012
Grasslands, forests, heathlands, and wetlands		N reduction	Recovery from N deposition likely to be a slow process		Review paper	Stevens, 2016
			Indicators of elevated N deposition and CL exceedance described and evaluated			Sutton *et al.*, 2001; Sutton *et al.*, 2004
Forests	Europe	Atmospheric N deposition	Slight shift towards nitrofylic species at high N deposition		Natural background variation may have overshadowed deposition effects due to large geographic range	van Dobben and de Vries, 2010
Saltmarshes			Increases vegetation growth			Van Wijnen and Bakker, 1999
Old sessile oak woodlands	Ireland	N deposition	• Increase in nitrophiles with higher N deposition • Community change point of 13.2 kg N ha^{-1} yr^{-1} recommended	Indicator species: *Quercus petraea-Luzula sylvatica*	1667 10 m × 10 m releves Total N deposition not correlated with soil pH	Wilkins and Aherne, 2016

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Northern hardwood forests (Sugar Maple dominant)	Great Lakes, North America	Wet chemistry and Fourier transform infrared spectroscopy (FTIR)	Decomposition is inhibited by N enrichment	Concentration of acid-insoluble fraction (AIF)	3-yr study	Xia *et al.*, 2017
Cinnamomum Camphora and *Pinus massoniana*	Guiyang,SW China	N concentrations and $\delta^{15}N$ values in soils and collected leaves to see feasibility of tall arbors as bio-indicators.	$\delta^{15}N$ values in leaves found to be lower than those of soil. $\delta^{15}N$ values of Masson pine leaves lower than camphor leaves.		Linear increase in N in leaves with atmospheric N deposition. Observed differences between urban and rural areas showing different N inputs. Higher than critical load.	Xu *et al.*, 2017
Water						
Freshwaters		Acidity due to N and decrease in S	Increases dissolved organic carbon (DOC) concentrations (causing brown colouring)			Butterbach-Bahl and Dannenmann, 2011
Freshwaters		Acidification of freshwater	Loss or damage to fish populations. Fish reproductive capability include effects on egg production, hatching success and on physiological parameters such as osmo-regulation and gill function			Donaghy and Verspoor, 1997; Kroglund *et al.*, 2007
					Review paper on critical loads and dose-response relationships. Includes several tables on exceedance indications	de Vries *et al.*, 2010
	Norway		Health of Atlantic salmon in rivers. Health of brown trout in lakes.			Lien *et al.*, 1996; Lydersen *et al.*, 2004; Kroglund *et al.*, 2007

Table 2. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Freshwater		Acidification	Changes in changes in epilithic diatoms, aquatic macrophytes, macroinvertebrate communities, and fish and bird populations			Ormerod and Durance, 2009
**Fresh water and coastal regions			Noxious and toxic algal blooms, increased turbidity, and disruption of ecosystem functioning, shifts in food webs and loss of fish stocks		Review paper	Rabalais, 2002
		Eutrophication	Alteration of aquatic food-webs and fish populations			Smith and Schindler, 2009
Freshwaters		Winter nitrate concentrations	Reduced macrophyte species richness			Van der Molen *et al.*, 1998
N limited lakes and Freshwaters		N deposition	High algal growth			Van der Molen *et al.*, 1998; Camargo and Alonso, 2006
Biodiversity						
Butterflies (grassland, heathland, woodland, and farmland)	The Netherlands	N deposition	• Induces excess early growth of plants • Changes quality of food available for larvae • Reduction in butterfly biodiversity quality	Butterflies as indicators	Index of butterfly sensitivity to N created (CNI) 17 yrs of data	Feest *et al.*, 2014
Lizards and heathlands			• Lack of management causes open bare ground unsuitable for habitat		Response to Maes *et al.*, 2017	Jones *et al.*, 2017
Lizards and heathlands			• Management effects detrimental to lizards		Comment on Jones *et al.* paper	Maes *et al.*, 2017
Non-calcareous dune grasslands	Ameland and Terschelling, the Netherlands	Increased N deposition causing encroachment	Encroachment of *Calamagrotis* caused severe decline in carabids		Effect of grazed and non-grazed grassland on carabid assemblages	Nijssen *et al.*, 2001
**Fauna		Increased N deposition	Direct and indirect effects on fauna		• Review paper • Scientific evidence on causal relationship between N deposition-fauna insufficient	Nijssen *et al.*, 2017

Table 2. *(continued)*.

Ecosystem Type/Species	Location	Approach	Ecological Impact	Effects Indicator	Comments	Reference
Heathlands and invertebrates (Diptera and carabid beetles)	The Netherlands	Soil chemistry, plant chemistry, fauna sampling	• Vegetation N:P ratio negatively linked to invertebrate density and species richness • Found to impact fauna more strongly than vegetation structure and plant species richness • Soil acidity and P-availability also main causes for loss of habitat structure and plant species richness	Vegetation N:P ratio	• Suggest importance of biogeochemistry and ecological stoichiometry	Vogels *et al.*, 2016
Butterflies (56 resident species)	The Netherlands	N deposition	• Butterfly species traits strongly dependent on N availability	Butterflies and CNI		WallisDeVries and van Swaay, 2016

(Bobbink *et al.*, 2010; Lu *et al.*, 2010; van Dobben and de Vries, 2010; Fenn *et al.*, 2011; Lu *et al.*, 2011; Lu *et al.*, 2014; de Vries *et al.*, 2015; Wilkins and Aherne, 2016; Du, 2017; Xia *et al.*, 2017). In sand dunes, effects include decalcification of topsoil, grass encroachment, changes in seed germination, and changes in species richness and composition (Ovesen, 2001; Jones *et al.*, 2004; Field *et al.*, 2014; Aggenbach *et al.*, 2016; Bird and Choi, 2016; Kooijman *et al.*, 2016).

Impacts from N enrichment on aquatic ecosystems include increased dissolved organic carbon (DOC) concentrations (Butterbach-Bahl and Dannenmann, 2011), loss or damage to fish populations due to changes in the capabilities in fish to reproduce (Donaghy and Verspoor, 1997; Kroglund *et al.*, 2008), and impacts to the health of fish in rivers (Atlantic salmon) and lakes (brown trout) (Lien *et al.*, 1996; Lydersen *et al.*, 2004; Kroglund *et al.*, 2008), changes in aquatic communities, such as epilithic diatoms, aquatic macrophytes, macroinvertebrates, and fish and bird populations (Van der Molen *et al.*, 1998; Ormerod and Durance, 2009; Smith and Schindler, 2009) and increase in algal growth (Van der Molen *et al.*, 1998; Camargo and Alonso, 2006; Hobbs *et al.*, 2010; Baron *et al.*, 2011; Fenn *et al.*, 2011).

Deposition of N on terrestrial and aquatic ecosystems has also resulted in direct and indirect effects on animal biodiversity due to changes in habitat structure and function (Nijssen *et al.*, 2001; Feest *et al.*, 2014; Jones *et al.*, 2016; Vogels *et al.*, 2016; Maes *et al.*, 2017), including declines in carabids and dipterals (Nijssen *et al.*, 2001; Vogels *et al.*, 2016), changes in habitats for lizards (Jones *et al.*, 2016), infestations of heather beetles (Berdowski and Zeilinga, 1983; Berdowski, 1993), and changes in butterfly species traits (Feest *et al.*, 2014; WallisDeVries and van Swaay, 2016). Other reported impacts on fauna due to N deposition include changes in food diversity, quality, and abundance, as well as decreased reproduction (Nijssen *et al.*, 2017).

Effects Indicators of Nitrogen Deposition

Impacts of nitrogen are widely assessed using the percent cover of individual plant species (Soons *et al.*, 2016) and the presence of an indicator species (Wilkins and Aherne, 2016). Useful plant indicators include the graminoid:forb ratio (Stevens *et al.*, 2006; Stevens *et al.*, 2009b) and epiphytic lichen and terricolous lichen taxa (Stevens *et al.*, 2012) with shifts in lichen communities occurring above a critical load of 5 kg N ha^{-1} yr^{-1} (Stevens *et al.*, 2012). Butterflies can be effective indicators for N deposition on faunal biodiversity, with an index of butterfly sensitivity to N having been created using 25 years of data based on changes in butterfly abundance (Feest *et al.*, 2014; WallisDeVries and van Swaay, 2016).

Tissue and litter N content and vegetation N:P ratio have been shown to be good biomonitors for shrubs and mosses and other organisms (Pitcairn *et al.*, 1998; Hicks *et al.*, 2000; Conti and Cecchetti, 2001; Pitcairn *et al.*, 2001; Pitcairn *et al.*, 2002; Pitcairn *et al.*, 2003; Mitchell *et al.*, 2004; Pitcairn *et al.*, 2006; McNeil *et al.*, 2007; Edmondson *et al.*, 2010; Caporn *et al.*, 2014; Harmens *et al.*, 2015; Rowe *et al.*, 2016; Vogels *et al.*, 2016; Du, 2017).

Table 3. Impacts of Acidifying Pollutant: Sulfur (S).

Ecosystem Type / Species	Location	Approach	Ecological Impact	Effects Indicators	Comments	Reference
Terrestrial ecosystems						
Ginkgoales, bennettites, and conifers	Astartekløft, East Greenland	Physiognomic leaf analysis on well-preserved fossil leaves	Modified leaf physiognomy (increase in leaf roundness)		Regular exposure to volcanic ash	Bacon *et al.*, 2013
Fruticose lichens (*Anaptychia ciliaris*, *Evernia prunastri,* and *Ramalina farinacea*)						Balaguer and Manrique, 1991
Pinus halepensis	Campi Flegrei, Southern Italy	Cuticle micromorphology on epicuticular and epistomatal waxes	Degradation of epicuticular and epistomatal waxes as fusion of the wax structures		Influence of volcanic gases. Method shows promise	Bartiromo *et al.*, 2012
			Aquatic and terrestrial ecosystems acidified			Doney *et al.*, 2007
Mesozoic gymnosperm and fern fossil floras	Growth chamber	SO_2 fumigation. Damage structures in leaf epidermis and cuticle	Smaller leaves that did not persist; distinct raised areas of cuticle surrounding stomata appeared; surface waxes altered; blistering of the cuticle occurred; and the stomatal complex became distorted		Volcanic SO_2. Method effective	Elliott-Kingston *et al.*, 2014
Picea abies and *Abies alba* tree cores	NE Italy	Synchrotron radiation				Fairchild *et al.*, 2009
Picea Abies	Sudetes, Southern Poland	Tree ring widths	Strongly negative dependence of tree ring widths on fog deposition rates. Acidification and tree dieback.		Study area was an ecological disaster site. Most trees >80 yrs old	Godek *et al.*, 2015
		SO_2 fumigation	Change in stomatal density and its ratio to the stomatal index			Haworth *et al.*, 2010; Haworth *et al.*, 2012
Cladia retipora (lichens)	Tasmania, Australia	Isotopic fingerprinting			Suitability for lichen bio-monitoring baseline determined ($\delta^{34}S_{mean} = 0.03 \pm 0.05‰$)	Hogan *et al.*, 2017
Forests	Czech Republic	Temporal and spatial trends	Spatial pattern change: 18.1–0.2 g m^{-2} yr^{-1} (1995–2011)		Based on long-term monitoring	Hůnová *et al.*, 2014
Grasses	Belgium	S fertilization	Yield increases			Mathot *et al.*,

Table 3. (*continued*).

Ecosystem Type / Species	Location	Approach	Ecological Impact	Effects Indicators	Comments	Reference
Grasses		Sulfur nutritional diagnostic tool on S fertilization			Indicator for predicting S deficiency using linear relationships	Mathot *et al.*, 2009
Industrial sources	Fraser Valley and Saturna Island, Canada	Isotopic mass balance approach			Heavily reduced H_2S from bacterial sulfate ($\delta^{34}S$ range = –1.6 – 9‰)	Norman *et al.*, 2004
Forests	Nacetin, Czech Republic	Throughfall sampling, water and soil sampling	Reversal of nitrogen enrichment of ecosystems, cessation of nitrate leaching, and a major loss of accumulated organic soil carbon and nitrogen stocks.			Oulehle *et al.*, 2011
Scots pine	Eurasia		Large scale decrease in growth; higher sensitivity to drought			Savva and Berninger, 2010
Grassland sites	UK (68 sites)	Measured surface and subsoil chemistry	Acidification from S deposition		Summers of 2002–2003	Stevens *et al.*, 2006
	Europe	S fertilization	Yield increases			Tarrasón *et al.*, 2006
Vegetation and soil	The Netherlands	Vegetation and soil sampling	Shift towards acidophytic species at high S-deposition			van Dobben and de Vries, 2010
Tree cores	NE Italy	Elemental analyser isotope ratio mass spectrometry (EA-IRMS) compared to tree rings				Wynn *et al.*, 2014
Cinnamomum camphora Camphor tree leaves	Shanghai, China	Synchrotron radiation X-ray fluorescence and X-ray absorption			Successfully used to study S concentration, speciation, and bioaccumulation	Zeng *et al.*, 2013

Table 4. Impacts of Base Cations.

Ecosystem Type/Species	Location	Approach	Ecological Impacts	Effects Indicator	Comments	Reference
Picea abies, Fagus sylvatica, and *Betula pendula*	South Sweden	Bulk deposition, throughfall, stemflow, litterfall, above-ground biomass increments and soil solution	• Net foliage leaching of Ca,Mg, and K from canopies • Net loss of base cations from soils, especially spruce		Soil acidification higher in spruce than in beach and birch	Bergkvist and Folkeson, 1995
Coniferous and deciduous forests	Northern Germany		• Substantial leaching of base cations from foliage			Bredemeier *et al.*, 1990
Leaf litter (*Alnus glutinosa, Acer pesudoplatanus* and *Fagus sylvatica*)	Vosges Mountains, NE France	Leaf litter decomposition response to acidification	Magnitude of inhibition of decomposition increases with increases in H^+ in Al concentration	Litter decomposition	Response stronger in coarse than in fine mesh bags	Ferreira and Guérold, 2017
Temperate, boreal, coastal, and alpine forests	Ontario, British Columbia, and Nova Scotia, Canada	STA to estimate base cation weathering rate		Soil silt content suggested as a better predictor of soil weathering rates	General model agreement with data	Koseva *et al.*, 2010
Forested catchments	SW China	Measurements and models	Simulations suggest decrease in soil base saturation and pH (soil degradation) over the last decades	Ca:S ratio		Larssen and Carmichael, 2000
Subtropical/tropical forest	Guangdong Province, China	Long-term N addition	• Significant soil acidification • Depleted base cations • Increase in exchangeable H^+ proportion in soil cation pools • Increase in soil CEC • Greatly decreased soil solution pH at 20 cm depth but not at 40 cm depth		No change in soil exchangeable Al^{3+}	Lu *et al.*, 2014
Northern forests	Quebec, Canada	Modelling soil chemical weathering rates			Changes in composition within soil profiles are mostly linear (degree of weathering, *k,* linear to calculated gain/loss of base cations)	Ouimet *et al.*, 2006; Ouimet, 2008

Table 4. (*continued*).

Ecosystem Type/Species	Location	Approach	Ecological Impacts	Effects Indicator	Comments	Reference
Mixed boreal forest (dominated by Norway spruce)	Eastern Finland	Sulphate deposition	Ca^{2+} and Mg^{2+} leaching with greater leaching during growing season tan dormant season	K^+ leaching higher than Ca^{2+} and Mg^{2+} leaching	3 yr study (Three 50 m × 50 m plots)	Piirainen *et al.*, 2002
Muskeg peatland, lakes, ponds, and uplands (jack pine and trembling aspen)	Oil sands region, Alberta, Canada	• Base cation weathering rates estimated • Deposition in throughfall and open collectors measured	Potential for soil acidification mitigated by high base cation deposition	(Ca + Mg):S ratio	63 sandy, acid-sensitive, upland forest sites	Whitfield *et al.*, 2011; Watmough *et al.*, 2014
Sugar maple	Northern Vermont	• Base cation depletion	Crown dieback in stands with low pH, low Ca and Mg, and high Al			Wilmot *et al.*, 1995

Empirical critical loads of N have been established for a range of terrestrial ecosystems, based on changes to ecosystem structure and function; a critical load of 15 kg N ha^{-1} yr^{-1} has been recommended for acidic and calcareous grassland (Bobbink and Hettelingh, 2011; Henry and Aherne, 2014), 15 kg N ha^{-1} yr^{-1} for calcareous grey dunes and 10 kg N ha^{-1} yr^{-1} for acidic grey dunes (Kooijman *et al.*, 2016). Similarly, critical levels for atmospheric N are 1 µg NH_3 m^{-3} for lichens and bryophytes and 3 µg NH_3 m^{-3} for herbaceous species (de Vries *et al.*, 2007; Cape *et al.*, 2009). de Vries *et al.* (2007) suggested critical N concentrations in soil solution for a variety of vegetation types to protect against N leaching, with concentrations ranging from 0.2 mg N L^{-1} for lichens and cranberries to 6.5 mg N L^{-1} for deciduous forests. Similarly, a general threshold for terrestrial soil C:N ratio of < 20–25:1 has been suggested as a threshold for nitrate leaching (Bähring *et al.*, 2017). Litter N content has also been found to be an effective indicator (White *et al.*, 1996; Pilkington *et al.*, 2005). Bähring *et al.* (2017) found shoot increment to be a more sensitive indicator than soil C:N ratio. Soil pH thresholds are suggested at 3.8 for forbs and 4.5 for acidic grasslands (Field *et al.*, 2014). Polyphenol oxidase (PPO) activity is also another biomarker of N enrichment with little difference above an application of 60 kg ha^{-1} yr^{-1} on peach trees (Edmondson *et al.*, 2010; Falguera *et al.*, 2012; Caporn *et al.*, 2014).

Indicators in aquatic ecosystems include shifts in the relative abundance of diatom communities, with a threshold for wet N deposition between 1.0 and 1.5 kg N ha^{-1} yr^{-1} (Baron, 2006; Saros *et al.*, 2011; Nanus *et al.*, 2012; Sheibley *et al.*, 2014); nitrate concentrations, where a threshold of 15 mg L^{-1} has been determined for well water (Panno *et al.*, 2006); and the ratio of dissolved inorganic N to total phosphorus (DIN:TP), with a suggested range between 1.5 and 3.4 (Bergström, 2010; Fenn *et al.*, 2011; de Vries *et al.*, 2015).

PAH DEPOSITION

Major PAH Species

The major PAH species are generally considered to be the US EPA's 16 PAH priority pollutants, which includes naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, and benzo[ghi]perylene. These PAH species were selected as priority pollutants because they are found in the environment, analytical standards and toxicity information are readily available, and many scientific investigations have focused on these 16 PAHs (Andersson and Achten, 2015). However, recent advances in analytical methods have broadened the range of analyses to include polycyclic aromatic compounds (PACs). PACs consists of the 16 PAHs as well as alkylated PAHs, unsubstituted and alkylated dibenzothiophenes, heterocyclic aromatic compounds, and PAC transformation products, which together comprise thousands of compounds

(Manzano *et al.*, 2017; Wnorowski and Charland, 2017). Although these additional PAC species have been the subject of far fewer studies relative to unsubstituted PAHs, their impacts have been observed in air, water, and wildlife particularly near oil sands mining regions (Kelly *et al.*, 2009; Lundin *et al.*, 2015; Schuster *et al.*, 2015; Zhang *et al.*, 2015; Jariyasopit *et al.*, 2016). Toxicity information on these compounds is still very limited; therefore, the following sections will focus on the impacts and biomonitors of PAHs.

Impacts of PAH Deposition

Impacts of PAH deposition on terrestrial ecosystems include visible leaf damage, chlorosis and necrosis, mesophyll collapse, leaf surface bronzing and silvering (Oguntimehin *et al.*, 2008; Oguntimehin *et al.*, 2010); oxidative stress response (Liu *et al.*, 2009); changes in biomass production (Wild *et al.*, 1992; Kummerová and Kmentová, 2004; Wieczorek and Wieczorek, 2007; Váňová *et al.*, 2009; Ahammed *et al.*, 2012a, b); changes in carbon allocation (Desalme *et al.*, 2011); changes to the soil-plant-rhizosphere system, such as enhancement by arbuscular mycorrhizal fungi and degradation by ectomycorrhiza (Koivula *et al.*, 2004; Joner *et al.*, 2006; Verdin *et al.*, 2006); and negative effects on photosynthesis (Wieczorek and Wieczorek, 2007; Oguntimehin *et al.*, 2008; Oguntimehin *et al.*, 2010; Desalme *et al.*, 2011; Ahammed *et al.*, 2012a, b).

Other effects of PAHs on living organisms include the enrichment of antibiotic resistance bacteria (ARB) in soils (Chen *et al.*, 2017) and liver tumours in aquatic organisms, for example, liver lesions in English sole, mortality of sensitive aquatic invertebrates, and skin ulcers and fin erosion in cod and corkwing wrasse (Aas *et al.*, 2001). In both humans and animals, PAHs have been found to be carcinogenic, mutagenic, teratogenic, and immunosuppressive (Malmgren *et al.*, 1952; Yasuhira, 1964; Philips *et al.*, 1973; Grimmer *et al.*, 1983; Blanton *et al.*, 1986; Hahon and Booth, 1986; Blanton *et al.*, 1988; Zhao *et al.*, 1990; Dasgupta and Lahiri, 1992; Zhao *et al.*, 1992; ATSDR, 1993; Szczeklik *et al.*, 1994; Boström *et al.*, 2002; Yan *et al.*, 2004; Satya *et al.*, 2012; Rodríguez-Estival *et al.*, 2015; Mętrak *et al.*, 2016) given their potential to form carcinogenic, mutagenic diols and epoxides that react with DNA (Shukla and Upreti, 2009).

Effects Indicators of PAH Deposition

Lichens have been shown to be effective indicators. Levels in lichens represent exposure (deposition) but there is no level associated with negative impacts of PAH deposition. Accumulation of PAH absorption in lichens results in PAH profiles that provide knowledge of both the origin and proximity of the source of the contamination (Migaszewski *et al.*, 2002; Blasco et al., 2006, 2008; Augusto *et al.*, 2009; Shukla and Upreti, 2009; Augusto *et al.*, 2010; Satya *et al.*, 2012; Shukla *et al.*, 2012; Augusto *et al.*, 2013). It should be noted that an indicator refers to qualitative data about an organism (e.g., changes in presence/absence, physiology), whereas a biomonitor includes both quantitative and qualitative data on exposure (deposition). Two other highly used biomonitors are mosses (Ötvös *et al.*, 2004; Wang *et al.*, 2009) and pine needles (Holoubek *et al.*, 2000; Conti and Cecchetti, 2001; Onianwa, 2001; Librando *et al.*, 2002; Migaszewski *et al.*, 2002; Piccardo *et al.*, 2005; Schröter-Kermani *et al.*, 2006; Yang *et al.*, 2007; Hwang and Wade, 2008; Lehndorff and Schwark, 2009; Augusto *et al.*, 2010; Ratola *et al.*, 2010; Sun *et al.*, 2010; Amigo *et al.*, 2011; Ratola *et al.*, 2011, 2012; Tomashuk *et al.*, 2012; Fernández-Varela *et al.*, 2015), with pine needles showing promise as better biomonitors for airborne PAH concentrations and mosses for atmospheric deposition (Oishi, 2013). The high PAH accumulation potential in wild rosemary leaves suggests this species to likely be a more valuable indicator than birch leaves or Scots pine needles, as found in a peat bog comparison study (Mętrak *et al.*, 2016). An effect, known as the forest filter effect, has been proposed to suggest that forests, such as deciduous forest canopies in Canada and Germany, intercept and transfer atmospheric PAHs to the surface (Su *et al.*, 2007). This effect shows the potential for using deciduous forests as biomonitors of PAH deposition. Other terrestrial ecosystem indicators include *Quercus ilex* L. Mediterranean evergreen oak (Librando *et al.*, 2002; De Nicola *et al.*, 2005; Orecchio, 2007; De Nicola *et al.*, 2008, 2011; Papa *et al.*, 2012; Baldantoni *et al.*, 2014), and to a lesser extent *Olea europaea* L. leaves *(*Baldantoni *et al.*, 2014); *Laurus nobilis* evergreen (Lodovici *et al.*, 1998); *Rhododendron oomurasaki* Azalea (Nakajima *et al.*, 1995); *Ficus benghalensis* leaves (Prajapati and Tripathi, 2008); *Calotropis gigantean* R. Br. leaves (Sharma and Tripathi, 2009); *Plantago* species (Bakker *et al.*, 1999; Bakker *et al.*, 2001); vascular plants (Desalme *et al.*, 2013); and the inner wood and rings of blue spruce trees (Rauert *et al.*, 2017). Biological indicators in animals include moose, grey wolf, and woodland caribou excrement (Lundin *et al.*, 2015) and non-lethal fecal sampling and muscle sampling of nestling tree swallows in the Athabasca Oil Sands (Cruz-Martinez *et al.*, 2015; Fernie *et al.*, 2018). Recent research has shown the potential of a passive dry deposition (PAS-DD) air sampling collector, composed of two parallel plates and a PUF disk, that can be used in the field as a replacement to biological indicators to monitor PAH deposition (Harner *et al.*, 2013; Eng *et al.*, 2014; Jariyasopit *et al.*, 2018).

In aquatic ecosystems, aquatic biomonitors include plants such as *Ceratophyllum demersum L.* and *Typha domingenesis pers* (Hassan *et al.*, 2010); sentinel zooplankton, e.g., Daphnia (Kurek *et al.*, 2013); spider webs (Rybak and Olejniczak, 2014); fish and invertebrates (Rose *et al.*, 2012); and crabs, such as spider crabs, snow crabs (Hellou *et al.*, 1994), or fresh water crabs *Sesarma boulengeri* (Salman *et al.*, 2014); and colonial waterbird eggs (Hebert *et al.*, 2011). Biliary fluorescence and DNA adducts have been found to be useful biomonitors for PAH contamination in cod and corkwing wrasse (Aas *et al.*, 1998; Aas *et al.*, 2000; Aas *et al.*, 2001; Hanson and Larsson, 2008), as well as in perch, northern pike, European eels, plaice, blue mussels, flounder, jackfish, and other fish species (Lin *et al.*, 1996; Ericson *et al.*, 1998; Richardson *et al.*, 2001; Ruddock *et al.*, 2003; Richardson *et al.*, 2004; Schiedek *et al.*, 2006; Vuorinen *et al.*, 2006; Beyer *et al.*, 2010; Nkpaa *et al.*, 2013; Ohiozebau *et al.*, 2017).

MERCURY DEPOSITION

Major Hg Species

There are three forms of atmospheric Hg including gaseous elemental Hg (GEM), gaseous oxidized Hg (GOM), and particulate-bound Hg (PBM). GEM is the dominant form of atmospheric Hg and capable of long range transport with an atmospheric lifetime of 0.5–1 year (Driscoll *et al.*, 2013). GOM and PBM typically have atmospheric lifetimes ranging from hours to weeks (Cole *et al.*, 2014). Hg is bi-directional in nature, with both emission and deposition occurring between the air and the surface (Wright *et al.*, 2016). All three forms of Hg can undergo dry deposition (Wright *et al.*, 2016); however, only GOM and PBM can also undergo wet deposition since oxidized Hg is more water soluble than GEM.

Impacts of Mercury Deposition

Mercury deposited to the Earth can undergo methylation in the presence of sulphate-reducing bacteria (SRB). During this process, oxidized Hg is converted to methylmercury (MeHg), which is more often referred to as monomethylmercury (MMHg) (Gilmour *et al.*, 1992; King *et al.*, 2000). Methylmercury bioaccumulates in biota and is highly toxic. Hg deposition impacts humans indirectly through the consumption of contaminated fish, wildlife, and plants that are contaminated with Hg (Meili, 1997; Meili *et al.*, 2003). The resulting effects are risks to the neurological, immune, and reproductive systems (Harada, 1995; Takeuchi *et al.*, 1996; Myers *et al.*, 1998; Schoeman *et al.*, 2009; Bose-O'Reilly *et al.*, 2010; Fernandes Azevedo *et al.*, 2012; Hong *et al.*, 2012; Rice *et al.*, 2014). Wildlife at highest risk are large predatory fish, fish-eating mammals, and fish-eating birds. Impacts on wildlife include reduced reproduction, changes to egg incubation times, behavioural changes, and neurological problems (Myers *et al.*, 1998; Wolfe *et al.*, 1998; Evers *et al.*, 2005; Scheuhammer *et al.*, 2007; Richard Pilsner *et al.*, 2010; Penglase *et al.*, 2014). The consumption of fish with high levels of Hg is an indirect impact of Hg deposition through bioaccumulation of MeHg in the fish and biomagnification in the food chain. Aquatic ecosystems that are acidic, nutrient-deficient, or have higher levels of dissolved organic matter (DOM) have higher concentrations of MeHg than their counterparts (Gilmour *et al.*, 1992; Watras and Huckabee, 1994; Driscoll *et al.*, 2013; French *et al.*, 2014). The effects of Hg on terrestrial and wildlife ecosystems are fairly well understood (Meili *et al.*, 2003; Evers *et al.*, 2005; Evers *et al.*, 2008; Caldwell *et al.*, 2009). Mercury concentrations in Canada are higher in the eastern part of the country than the west owing to the legacy of high emission sources to the south, the preponderance of acidic lakes and soils (Depew *et al.*, 2013, and references therein), and the retention of Hg in organic soils in lake catchments in eastern Canada (Dennis *et al.*, 2005).

Effects Indicators of Mercury Deposition

Ecosystem indicators that can exacerbate Hg bioaccumulation are total phosphorus (TP), DOC, acid status, and pH. Critical loads for Hg (and persistent organic pollutants; POPs) are not as well developed as they are for N and S. The concentration threshold used as an indicator of Hg sensitivity for total phosphorus is 0.03 mg L^{-1}, with fish having higher Hg below that threshold. DOC indicator values for Arctic tundra lakes range from 4 mg L^{-1} to 8 mg C L^{-1} (Driscoll *et al.*, 2007; French *et al.*, 2014; Stoken *et al.*, 2016). Ecosystem attributes which are associated with higher concentrations of bioavailable Hg include an ANC less than 100 µeq L^{-1}, and a pH less than 6.0. The critical limit for Hg concentration in drinking water is 6 µg L^{-1} (WHO, 2011). The Canadian water quality guidelines for the protection of aquatic life are 26 ng L^{-1} for inorganic Hg in freshwater, 16 ng L^{-1} in marine surface waters and 4 ng L^{-1} for MeHg in freshwater (CCME, 2003).

Suitable aquatic Hg biomonitors are walleye and northern pike, with thresholds of 0.16 ppm in prey fish and 0.3 ppm in fish tissue (Simoneau *et al.*, 2005; Drevnick *et al.*, 2007; Kinghorn *et al.*, 2007; Rasmussen *et al.*, 2007; Iwu, 2010; Monson *et al.*, 2011; Åkerblom *et al.*, 2012; Gandhi *et al.*, 2015). A good wildlife Hg biomonitor species is the common loon, where the threshold in loon blood is 3.0 ppm (Evers *et al.*, 2003; Kenow *et al.*, 2003; Burgess *et al.*, 2005; Champoux *et al.*, 2006; Evers *et al.*, 2008). Omnivorous birds, such as the Greylag goose, have been found to have higher levels of Hg in their feathers than birds with other dietary habits (Ahmadpour *et al.*, 2016). Mercury levels in marine birds have been studied extensively (Monteiro and Furness, 1995; Bearhop *et al.*, 2000; Becker *et al.*, 2002; Goodale *et al.*, 2008; Burger *et al.*, 2009; Ackerman *et al.*, 2014; Mashroofeh *et al.*, 2015; Binkowski *et al.*, 2016) with indicator thresholds determined to be 1.3 ppm in the blood of songbirds, and 10.0 ppm in the hair of bats. Hg in colonial waterbird eggs and wood frogs has also been measured (Hebert *et al.*, 2011, 2013; Abbasi *et al.*, 2015). In Canada, the Hg limit is 0.2 ppm for safe fish consumption, whereas elsewhere it is 0.5 ppm (de Vries *et al.*, 2007; de Vries *et al.*, 2015). Hg levels in human urine, blood, and hair can also be used as indicators (Sherlock *et al.*, 1984; Apostoli *et al.*, 2002; Myers *et al.*, 2003; Schober *et al.*, 2003; McDowell *et al.*, 2004; Caldwell *et al.*, 2009).

TRACE METAL DEPOSITION

Major Trace Metal Species

The 13 major trace metal species considered as priority pollutants by US EPA are Antimony (Sb), Arsenic (As), Beryllium (Be), Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), Mercury (Hg), Nickel (Ni), Selenium (Se), Silver (Ag), Thallium (Tl) and Zinc (Zn). These metals are part of the priority pollutants list because of their toxic effects on biota and occurrence in the environment. They also have chemical standards and analytical methods available and are produced in significant quantities (US EPA, 2017).

Impacts of Trace Metal Deposition

The impacts of trace metal deposition are well documented but especially for Pb. Much of the knowledge of the impacts of trace metals such as aluminum, vanadium, and nickel to terrestrial ecosystems has come from acidification studies as decreased pH mobilises metals causing changes that include changes to the mycorrhiza and fine root systems of plants, chlorosis, dwarfing, and reduced root and shoot growth (Rosseland *et al.*, 1990; Efroymson *et al.*, 1997; Ewais, 1997; Wang and Liu, 1999; Vachirapatama *et al.*, 2011).

Trace metals can accumulate in the suspended particulates and sediments in aquatic ecosystems causing serious potential risks to the health of ecosystems (Li *et al.*, 2013). Impacts of trace metals to aquatic ecosystems include effects on gill function by copper (Sola *et al.*, 1995; Rajkowska and Protasowicki, 2013), iron, manganese, and zinc (Rajkowska and Protasowicki, 2013); nervous systems by zinc, manganese, and iron (Baatrup, 1991; Takeda *et al.*, 2004); and growth and reproduction rates by lead, mercury, and cadmium (Mance, 1987; Ebrahimi and Taherianfard, 2011). Aluminum has been found to be toxic to fish and invertebrates and can interfere with the metabolic, reproductive, and breathing processes of mammals and birds, with potential links to tumours (Rosseland *et al.*, 1990; Exley *et al.*, 1991; Bast, 1993; Sparling and Lowe, 1996; Slaninova *et al.*, 2014). Cadmium has been linked to changes in growth and reproduction of biota (Eisler, 1985; Gallego *et al.*, 2012) and earthworms (Will and Suter, 1995; Chen *et al.*, 2017). Nickel accumulation has been linked to changes in metabolism, bone densities, growth, and survival in birds (Outridge and Scheuhammer, 1993; Eisler, 1998). Histopathological changes and other effects have been observed in carp due to trace metals (Vinodhini and Narayanan, 2009; Georgieva *et al.*, 2014). In humans, trace metal toxicities include damage to the lungs and nervous systems by Al; damage to the lungs, kidneys, and bones and renal dysfunction by Cd; and damage to the lungs, nasal passages, and skin by Ni (Williams and Burson, 1985; Denkhaus and Salnikow, 2002; Krewski *et al.*, 2007; Trzcinka-Ochocka *et al.*, 2010; Zambelli and Ciurli, 2013).

Effects Indicators of Trace Metal Deposition

Terrestrial biomonitors of trace metals include the use of mycorrhyzal fungi (Leyval *et al.*, 1997); *Quercus ilex* L. (Papa *et al.*, 2012); lichens (Garty, 2000; Augusto *et al.*, 2004; Augusto *et al.*, 2007; Augusto *et al.*, 2009); mosses and pine needles (Glooschenko, 1989; Bargagli *et al.*, 1995; Steinnes, 1995; Figueira *et al.*, 2002; Migaszewski *et al.*, 2002; Schilling and Lehman, 2002; Schintu *et al.*, 2005; Shotyk *et al.*, 2014; Singh *et al.*, 2017); bryophytes (Nimis *et al.*, 2002); *taraxacum officinale* dandelions (Kabata-Pendias and Dudka, 1991; Królak, 2003); *Tanacetum vulgare* tansy (Jasion *et al.*, 2013); and seagrass leaves (Govers *et al.*, 2014). Other biomonitors of metal contamination include metal enrichment factors and the geoaccumulation index (Abrahim and Parker, 2008; Çevik *et al.*, 2009; Zhang *et al.*, 2009; Lourino-Cabana *et al.*, 2011). Large-scale biomonitoring surveys of trace metal concentrations in moss tissue are carried out across Europe (Eurasia) on a five-year cycle under ICP Vegetation (ICP Vegetation, 2017).

In aquatic ecosystems and wildlife, levels of trace metals have been measured and used as indicators in the eggs, feathers, and tissues of Canadian geese (Tsipoura *et al.*, 2011); *Parus major and Ficedula hypoleuca* bird excrement (Berglund *et al.*, 2015); *Parus major and Carduelis sinica* birds (Deng *et al.*, 2007); harp seals (Agusa *et al.*, 2011); *Perna viridus* green-lipped mussels (Phillips, 1985); California mussels (Gordon *et al.*, 1980); Perna perna mussels (Bellotto and Miekeley, 2007); several fish species (Jaffar *et al.*, 1995; Atobatele and Olutona, 2015); earthworms (Morgan and Morgan, 1988; Stürzenbaum *et al.*, 2004; Hirano and Tamae, 2011; Chen *et al.*, 2017); and sentinel terrestrial animals, such as deer mice and meadow voles (Rodríguez-Estival and Smits, 2016). Critical limits for dissolved trace metal concentrations in drinking water include 3 $\mu g\ L^{-1}$ for Cd, 10 $\mu g\ L^{-1}$ for Pb, 2000 $\mu g\ L^{-1}$ for Cu, and 0.0078 $\mu g\ L^{-1}$ for Zn (de Vries *et al.*, 2007; WHO, 2011).

OZONE DEPOSITION

Impacts of Ozone Deposition

The impact of high ozone concentration on vegetation was traditionally considered as an exposure effect (concentration-based approach) (Fuhrer *et al.*, 1997). In recent decades, the accumulated stomatal flux (dose-based approach) was considered to be more suitable because ozone damage to vegetation is caused by injury to internal plant issue and subsequent reduction in photosynthesis, plant growth, and productivity (Musselman *et al.*, 1994; WHO, 2000; Filella *et al.*, 2005; Ferretti *et al.*, 2007). An example of the comparison of the different approaches was shown in Zhang *et al.* (2006). This section focuses on effects of O_3 deposition on vegetation. Ozone is a highly reactive oxygen species and impacts human and animal health, as well as terrestrial ecosystems (Chappelka and Samuelson, 1998; Ashmore, 2005; Ashmore *et al.*, 2006; Emberson and Buker, 2011; Mills *et al.*, 2011; Fuhrer *et al.*, 2016; Bergmann *et al.*, 2017, and references therein). In mammals, significant impacts on respiratory health include reductions in lung function and changes in lung structure (Mckee and Rodriguez, 1993; Chen *et al.*, 2007; Poursafa *et al.*, 2011; Bergmann *et al.*, 2017). Impacts to terrestrial ecosystems include decreased net photosynthesis in trees (Wittig *et al.*, 2009), decreased tree biomass (Karlssen *et al.*, 2003; Wittig *et al.*, 2009), decreased grassland productivity (Volk *et al.*, 2006), reduced flowering and bulb growth in woodland ground flora species (Keelan, 2007), plant cell death (Vainonen and Kangasjärvi, 2015), reduced cover of grass species (Thwaites *et al.*, 2006), decreased yields of rice and winter wheat (Feng *et al.*, 2003), agricultural yield losses (Van Dingenen *et al.*, 2009), changes to litter (Fuhrer *et al.*, 2016), changes in the decomposition rate of litter (Lindroth, 2010), among many others (Matyssek and Sandermann Jr., 2003). Rather than provide a table for O_3, the reader is referred to an extensive review provided by Bergmann *et al.* (2017) that examines over 450 references of plant exposure to O_3 in literature. In addition, a review of impacts of O_3 on terrestrial biodiversity and their downstream effects highlights the aboveground and belowground effects of O_3 exposure (Fuhrer *et al.*, 2016).

Effects Indicators of Ozone Deposition

Biomonitors for ozone have been studied intensively using field studies, such as the high use of white clover in ozone gardens (Villányi *et al.*, 2008; ICP Vegetation, 2017). There are, however, sometimes difficulties in distinguishing between the impacts being due to O_3 or other pollutants, such as reactive N (Fuhrer *et al.*, 2016). Useful biomonitors for O_3 deposition include the highly-sensitive Myrtaceae and Salicaceae families (Bergmann *et al.*, 2017) and whereas lichens, on the other hand, are not effective biomonitors due to their high O_3 tolerance (Bertuzzi *et al.*, 2013). An interesting ranking of native herbaceous and woody plant species can be found in Bergmann *et al.* (2017). Visible indicators on plants have been found to be leaf browning (Panigada *et al.*, 2009) and necrotic lesions on leaf surfaces, such as on the upper leaf surfaces of tobacco cultivars (Ashmore *et al.*, 1978; Ribas *et al.*, 1998; Klumpp *et al.*, 2006; Kafiatulla *et al.*, 2012; Vainonen and Kangasjärvi, 2015). Other non-visible biomarkers for plants that have been found to show potential are stress transcripts, proteins, and metabolites (Sandermann, 2000). In humans, systemic responses such as inflammation and oxidative stress have been found to be the most useful (Goodman *et al.*, 2015). Recently, an atmospheric critical level concentration for O_3 of 80 ppb was suggested for two Mediterranean trees, although this was performed under controlled conditions and has yet to be tested in the field (Fusaro *et al.*, 2017). Interestingly, in this study, N deposition was observed to counter the effects of O_3 deposition (Fusaro *et al.*, 2017).

CONCLUSIONS AND RECOMMENDATIONS

The ecosystem and human health impacts of the atmospheric deposition of various pollutants, and the indicators and biomonitors that have been found to be effective for these pollutants are summarized in this review paper. The impacts of the deposition of acidifying pollutants, eutrophying N, PAHs, Hg, Pb, and O_3 have been well-documented, whereas less is known about the impacts of alkylated and heterocyclic PAC and trace metals, such as iron and manganese. Further studies on possible biomonitors for PAC and O_3 are needed. Many studies have been tested under controlled conditions only. Future work should include testing suggested effects indicators, such as the critical load of O_3 on Mediterranean trees, in the field. The current understanding of effects and indicators is due to close collaboration between various research communities. This integration is essential to addressing environmental problems. For example, it would be interesting to explore the possibility of extending the use of the STressor-Ecological Production function-final ecosystem goods and Services (STEPS) Framework, which has been applied to aquatic eutrophication (Rhodes *et al.*, 2017), to other chemical species, such as PAHs, Hg, and trace metals. Recent success by Fernie *et al.* (2018) and Cruz-Martinez *et al.* (2015) on fecal and muscle sampling of PAHs and other contaminants in nesting tree swallows is being built upon with continuing research on toxicity in these birds. Perhaps expanding the list of chemical species, such as adding Hg, in these fecal samples could be performed concurrently to expand our knowledge of these wild birds and the effects of the Alberta Oil Sands on them and to develop this species as a potential biomonitor for future studies.

ACKNOWLEDGEMENTS

This work was supported by the Climate Change and Air Pollutants program of Environment and Climate Change Canada and by the Province of Alberta through the Oil Sands Monitoring Program.

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Received for review, March 25, 2018
Revised, June 20, 2018
Accepted, June 25, 2018