Review

Influence of surface chemistry of carbon materials on their interactions with inorganic nitrogen contaminants in soil and water

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HIGHLIGHTS

- Review of chemisorption of inorganic nitrogen contaminants (INC) on carbon surface.
- Carbon's surface chemistry plays a critical role in adsorption of INC.
- Oxygen containing surface functional groups have been found to be more influential.
- Basic sites on carbon surface influence chemisorption of anionic INC.
- pH and pHzpc and elemental and mineral composition of carbon are also important.

GRAPHICAL ABSTRACT
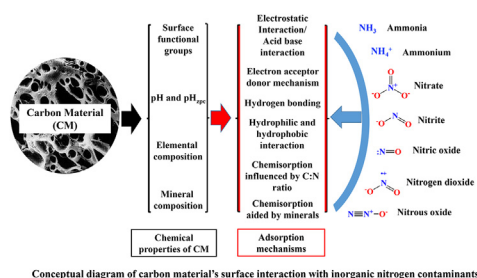
Conceptual diagram of carbon material's surface interaction with inorganic nitrogen contaminants

ARTICLE INFO

ABSTRACT

Inorganic nitrogen contaminants (INC) (NH_4^+, NO_3^-, NO_2^-, NH_3, NO, NO_2, and N_2O) pose a growing risk to the environment, and their remediation methods are highly sought after. Application of carbon materials (CM), such as biochar and activated carbon, to remediate INC from agricultural fields and wastewater treatment plants has gained a significant interest since past few years. Understanding the role of surface chemistry of CM in adsorption of various INC is highly critical to increase adsorption efficiency as well as to assess the long term impact of using these highly recalcitrant CM for remediation of INC. Critical reviews of adsorption studies related to INC have revealed that carbon surface chemistry (surface functional groups, pH, Eh, elemental composition, and mineral content) has significant influence on adsorption of INC. Compared to basic functional groups, oxygen containing surface functional groups have been found to be more influential for adsorption of INC. However, basic sites on carbon materials still play an important role in chemisorption of anionic INC. Apart from surface functional groups, pH, Eh and pH_{zpc} of CM and elemental and mineral composition of its surface are important properties capable of altering INC interactions with CM. This review summarizes our current understanding of INC interactions with CM's surface through the known chemisorption mechanisms: electrostatic interaction, hydrogen bonding, electron donor-acceptor mechanism, hydrophobic and hydrophilic interaction, chemisorption aided by minerals, and interactions influenced by pH and elemental composition. Change in surface chemistry of CM in soil during aging is also discussed.

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Nomenclature

AC	Activated carbon		EDAM	Electron donor-acceptor mechanism
AEC	Anion exchange capacity		Eh	Redox potential
ANAMMOX	Anaerobic ammonium oxidation		EI	Electrostatic interaction
BC	Biochar		HB	Hydrogen bonding
CAM	Chemisorption aided by minerals		HHI	Hydrophobic and hydrophilic interaction
CEC	Cation exchange capacity		INC	Inorganic nitrogen contaminants
CICN	Chemisorption influenced by C:N		OCSFG	Oxygen containing surface functional groups
CM	Carbon material		NOM	Natural Organic matter
DNRA	Dissimilatory nitrate reduction to ammonium		NPE	Negative priming effect
DOC	Dissolved organic carbon		PZNC/pH_{zpc}	Point of zero net charge/pH at zero point of charge
DOM	Dissolved organic matter		PPE	Positive priming effect
			SCR	Selective catalytic reduction
			SFG	Surface function groups

1. Introduction

Nitrogen (N) is a key element in biomolecules required for existence of life on Earth (Mochida et al., 2000; Philippot et al., 2007), but N in its inert gaseous form (N_2) cannot be directly utilized by organisms (Hoffman et al., 2014). It is converted to bioavailable, inorganic forms of N, NH_4^+ (ammonium) or NO_3^- (nitrate), via three important mechanisms: (i) by nitrogen fixing bacteria in soil (Butterbach-Bahl et al., 2013), (ii) industrially synthesized (Haber Bosch process) (Gu et al., 2013), and (iii) during lightning (Zahran, 1999). Apart from N demand by living organisms, N containing compounds are used in the manufacturing of fertilizers, explosives, dye intermediates, solvents, pharmaceuticals, and many other industries (Scott et al., 2014a).

Climate change, unsustainable agriculture practices (high N input), and disruption in natural hydrological flow processes has disrupted global N cycle (Hagedorn et al., 2001), subsequently affecting N distribution in atmospheric, terrestrial, and aquatic ecosystems (Galloway et al., 2003). Chemical fertilizers used in agriculture are the major source of inorganic nitrogen contaminants (INC) in soil which also enter surrounding water bodies through infiltration and surface runoffs. It is estimated that almost 50% of the fertilizer applied to soil is incorporated into crops while the rest is emitted to the atmosphere (NO, NO_2, N_2O, NH_3, or N_2) or released into water bodies (NH_4^+, NO_3^-, and NO_2^-) (Galloway et al., 2003).

Inorganic contaminants pose greater risk to the ecosystems since they are more stable in the environment (Beesley et al., 2011). High concentration of ammonia nitrogen (NH_3 and NH_4^+) leads to eutrophication, reduces oxygen concentration (Cheng et al., 2016), damages internal organs, and affects metabolic activities of aquatic organisms (Zhou et al., 2015). Ingestion of NO_3^- via drinking water, and its subsequent *in vivo* conversion to NO_2^-, causes methemoglobinaemia in infants, stomach cancer in adults, and other ailments in livestock (Loganathan et al., 2013). NO_x (NO_2 and NO) and N_2O emitted into the atmosphere causes acid rain (Cameron et al., 2013) and ozone depletion (Phillips et al., 2016), respectively. In addition, N_2O is a major greenhouse gas (Phillips et al., 2016).

There are various treatment methods available to remediate INC, and there is a growing interest in application of carbon materials (CMs), mainly activated carbon (AC) and biochar (BC), as effective adsorbents for removal of INC. CMs are used in various environmental management applications, including (i) soil remediation and improvement, (ii) waste recycle, and (iii) climate change mitigation (N_2O adsorption and C sequestration) (Ahmad et al., 2014; Ding et al., 2016; Tang et al., 2013; Woolf et al., 2010; Zhao et al., 2013). As per an estimate by Woolf et al. (2010), application of BC as a soil amendment can reduce global emissions by 1.8 Pg CO_2 equivalent annually and 130 Pg CO_2 equivalent over the next century, at the current rate of resources available for BC production.