

Perchlorate in an urban lawn environment

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Environmental context. The extent of perchlorate contamination in our environment is inadequately known. We examined perchlorate content in precipitation, soil water, soil cores and grass clippings in an urban lawn environment. Our results show that perchlorate is present in the lawn environment at concentrations that may adversely affect human health.

Abstract. Perchlorate contamination in groundwater is a concern owing to the likelihood that low concentrations of perchlorate may disrupt normal thyroid function. Lawns fertilized with perchlorate-containing fertilizers can adversely affect groundwater. We examined perchlorate concentrations from June 2006 to January 2007 in precipitation, grass clippings, soil cores and soil water at eight lawn environments in Suffolk County, NY, a county where lawns comprise more than 25% of the land use. Measured concentrations in soil waters were as high as $255 \mu\text{g L}^{-1}$, with average concentrations of $69 \mu\text{g L}^{-1}$ beneath sites treated with organic fertilizer, $1.2 \mu\text{g L}^{-1}$ beneath sites treated with chemical fertilizer and $0.34 \mu\text{g L}^{-1}$ beneath sites not fertilized. Although concentrations were dependent on the type of fertilizer applied, the patterns of leaching and grass uptake were similar at all sites.

Additional keywords: fertilizer, grass, groundwater, New York.

Introduction

Since perchlorate was added to the USA Environmental Protection Agency contaminant candidate list in 1998,^[1] the extent of perchlorate contamination has been the focus of much research and debate. Perchlorate is known to inhibit iodide uptake of the thyroid gland, which is of particular concern for women and children with iodide deficiencies. This health concern coupled with the mobility of perchlorate poses a threat to groundwater. Our study area, Suffolk County, Long Island, NY (Fig. 1), is particularly sensitive to groundwater contamination as all potable water is derived from the local aquifers. New York State has implemented state action levels of $18 \mu\text{g ClO}_4^- \text{L}^{-1}$ for the public

notification level and $5 \mu\text{g L}^{-1}$ for the drinking water planning level in groundwater. In October 2008, the US EPA made a preliminary decision not to regulate perchlorate, a decision that may not be warranted.^[2] Then, in December 2008, the US EPA set an interim health advisory level at $15 \mu\text{g L}^{-1}$ based on the reference dose recommended by the National Research Council.^[3]

Perchlorate contamination is commonly associated with rocket fuel propellant or Chilean nitrate use. Chilean nitrate was historically used in agricultural fertilizers before the Haber-Bosch process of ammonium production (~1930) and is currently an acceptable form of nitrate used in US Department of Agriculture-certified organic fertilizers, although not all organic

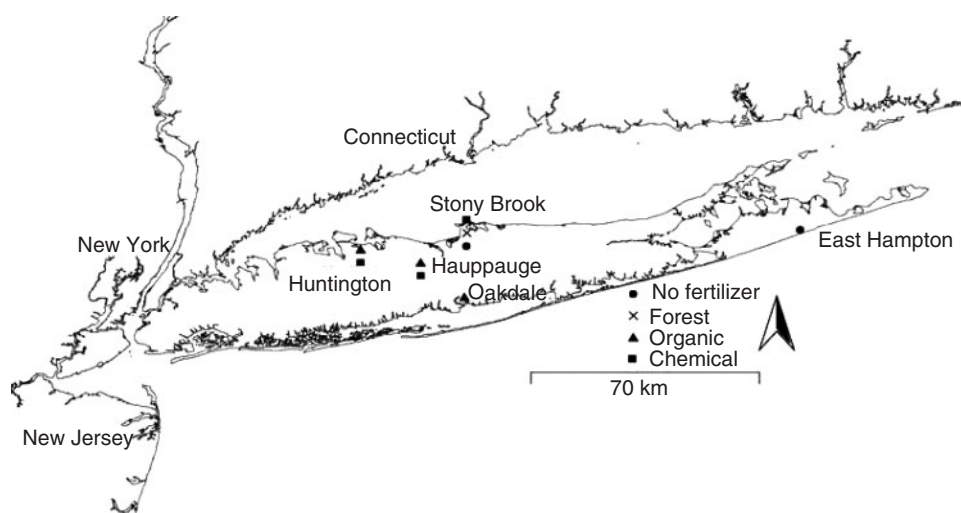


Fig. 1. Map of study locations. Most locations have multiple sites and all locations have a precipitation gauge.

fertilizers contain Chilean nitrate.^[4] Nitrate and presumably perchlorate have concentrated in the caliche ore through atmospheric deposition and concentration owing to low rainfall in the area.^[5-7] Perchlorate is rarely detected in fertilizer products that do not contain caliche ore.^[8,9] The amount of perchlorate in Chilean nitrate has varied over time, with values as high as 6.8%,^[10] decreasing recently to values between 0.15 and 0.18%.^[4] A modified refining process is currently producing Chilean nitrate containing 0.01% perchlorate.^[10] Even at low concentrations, perchlorate from organic fertilizers could adversely affect groundwater concentrations depending on the dilution of Chilean nitrate in the fertilizer, the application rate of the fertilizer, and the imposed standards for drinking water quality.

The fate of perchlorate from lawn fertilizers is uncertain. Studies^[8,9] conclude that lawn fertilizers are not used in great enough abundance to warrant concern for water quality. However, an increasing amount of land is being converted to turfgrass. In the United States, turfgrass is estimated to cover up to 14% of the total USA area, including residential, commercial and industrial lawns, parks, golf courses and athletic fields.^[11,12] A 1970s land-use survey concluded that turfgrass covers more than 25% of the 2300 km² that is Suffolk County, NY. This value has likely increased since then, as residential land use has increased. The soils in Suffolk County lack the needed nutrients for healthy turfgrass to grow; consequently, fertilizers are applied throughout the year. The local water authority is promoting an organic fertilizer campaign in hopes of reducing nitrate leaching to groundwater and to reduce water demands from irrigation that may, in fact, increase the amount of perchlorate applied to lawns.^[13]

Perchlorate applied to a lawn may either be taken up by the plants or leach to groundwater. Perchlorate uptake and bioconcentration in plant matter has been documented,^[14-18] although it is not certain why plants take up ClO₄⁻. Perchlorate retention in soil is negligible.^[19] Perchlorate concentrations in the unsaturated and saturated zones may decrease owing to microbial degradation depending on the type of microbial community, the exposure time of ClO₄⁻ in the environment, and the concentration of the competing ions, such as NO₃⁻.^[17,20,21] As NO₃⁻ is in much higher concentrations than ClO₄⁻ in a lawn environment, ClO₄⁻ degradation probably will not occur.

Our aim was to evaluate the concentrations of ClO₄⁻ leaching beneath maintained urban lawns in Suffolk County, NY. Here all potable water is derived from the local aquifers and the sandy nature of the soils makes it highly likely that soil water will quickly recharge the well-aerated aquifers. Soil water samples were collected monthly from suction lysimeters installed beneath lawns treated with organic fertilizer, chemical fertilizer or no fertilizer from June 2006 to January 2007. In addition, monthly samples of bulk precipitation and lawn clippings were collected and in November 2006, soil cores were collected at two sites treated with organic fertilizer. All samples were analysed for perchlorate, nitrate and chloride.

Experimental

Site descriptions

Five study locations were established in Suffolk County, NY, at maintained lawns at either the Stony Brook University Campus or at an office building site of the Suffolk County Water Authority. Most locations have multiple study sites (Fig. 1). Two of the sites were not fertilized, three were treated with organic fertilizer and three were treated with chemical fertilizer. There was also one site in a forested area at the University. (Only one sample, however, could be collected in the forested site owing to low sample volumes. This sample contained less than 0.1 µg L⁻¹ of ClO₄⁻ and will not be discussed further.) The Stony Brook site treated with chemical fertilizer and the site not fertilized were established in 2001. The Oakdale, Huntington and Hauppauge sites treated with organic fertilizer were established in 2002. The Huntington and Hauppauge sites treated with chemical fertilizer were established in 2005. The organic sites were fertilized with Pro-Grow fertilizer (North Country Organics, purchased from Bayles Garden Center and Nursery, Port Washington, NY), receiving 40 kg N ha⁻¹ year⁻¹ (Fig. 2). Pro Grow is composed of natural sulfate of potash, phosphate rock, colloidal phosphate, oyster meal, kelpmeal, greensand, vegetable and animal protein meals, natural nitrate of soda, compost and dried whey. Manure was not used. The chemical sites were fertilized with Scotts or Lesco Brand fertilizer (Home Depot, New York), receiving 150 or 200 kg N ha⁻¹ year⁻¹ (Fig. 2). All fertilizers were applied according to the recommendations of the manufacturers, so although the application rates vary, they are consistent

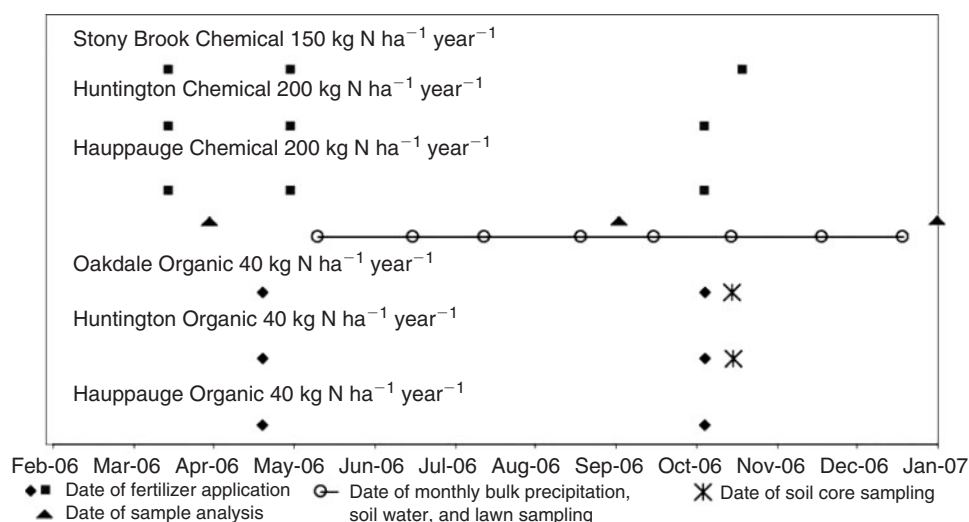


Fig. 2. Fertilizer and sampling time-line.

with what a fertilizer program at a home or business might be. All locations, except for Stony Brook, were irrigated with an automatic sprinkler system in the summer months. All lawns were mowed, once a week or once every other week, from April to November. The mowed grass was mulched and left on the lawns.

Sample collection and analysis

At each location, precipitation was collected monthly using All-Weather Rain Gauges purchased from Fisher Scientific online (<http://www.fishersci.com/wps/portal/HOME>, accessed 1 January 2009). These gauges sample both wet and dry deposition as they are not covered during dry periods. At each site, soil water samples were acquired from suction lysimeters collected monthly from June 2006 through January 2007. Ceramic suction lysimeters were installed in 2001, 2002 and 2005 at 100 cm below the surface. This depth was chosen because it is below the rooting depth of lawns and based on the assumption that the water at 100 cm will likely reach the groundwater table with minimal changes. Soil water and bulk precipitation samples were filtered in the field using a 0.2- μm surfactant-free cellulose acetate (SFCA) filter and collected in untreated polypropylene vials. Samples were stored in a cooler while in the field and then at 4°C until analysed.

Samples of the live grass were collected monthly from June 2006 through January 2007. A rectangular outline, 14 by 24.3 cm, was used so that a similar area of grass was collected each time. The grass was cut as close to the ground surface as possible, being sure not to sample soil, fallen leaves or mulched grass. Plants other than turfgrass, such as crabgrass, plantain, clover and dandelion were not excluded during sampling. Plant samples were brought back to the laboratory in brown paper bags and air-dried, then oven-dried at 105°C for 24 h. Samples weighing from 0.5 to 1.5 g were cut into small pieces and placed in 30 mL of deionised water in a 50-mL centrifuge tube and boiled in a water bath for approximately 1 h.^[14,17] The tubes were then placed in the refrigerator and manually shaken every

2 h for 8 h, then left overnight. The following day, the samples were centrifuged at $\sim 700g$ (2000 rpm on a 19-cm centrifuge at room temperature) for 30 min. The supernatant was filtered with a 0.45- μm glass fibre filter and then a 0.2- μm SFCA filter. Samples were stored at 4°C until analysed.

Two soil cores were collected at sites treated with organic fertilizer in November 2006, one at Hauppauge and one at Oakdale. The cores were sampled every 5 cm to a depth of ~ 100 cm using a combination of a soil auger and an Art's Manufacturing and Supply Inc. soil core sampler from Forestry Suppliers (Jackson, MS). Samples were first air-dried, then oven-dried at 110°C for 24 h. The soil was crushed using a shatterbox with an agate grinding bowl. Approximately 10 g of crushed soil was added to 10 mL of deionised water in a centrifuge tube and shaken vigorously by hand. The tubes were then centrifuged at $\sim 700g$ for 30 min. The supernatant was filtered with a 0.2- μm SFCA filter and diluted in deionised water with a 1 : 4 soil to water ratio.^[22]

All fertilizers analysed in the present study were prepared by dissolving ~ 5 g of fertilizer in 45 mL deionised water, vigorously shaking, then allowing the solution to sit for 1 h, centrifuging for 1 h, then filtering.^[8,9] All samples were fully dissolved before filtering. Solutions were diluted by 50 before analysis.

Holding times for our samples were not important because proper handling procedures were adhered to, i.e. filtering in the field and immediately storing at 4°C. Samples were analysed in April 2006, September 2006 and February 2007 (Fig. 2). Perchlorate samples were analysed using a sequential ion chromatography–mass spectroscopy–mass spectroscopy (IC-MS/MS) technique^[23] with a method detection limit of 0.1 $\mu\text{g L}^{-1}$ for soil water, grass and soil samples and a detection limit of 0.005 $\mu\text{g L}^{-1}$ for bulk precipitation samples. To account for matrix effects, all samples were spiked with an oxygen isotope (^{18}O)-labelled ClO_4^- internal standard. The precision determined from replicate analysis was $\pm 5\%$. Perchlorate in the deionised water was below the detection limit of 0.1 $\mu\text{g L}^{-1}$. Nitrogen as nitrate and chloride concentrations were analysed

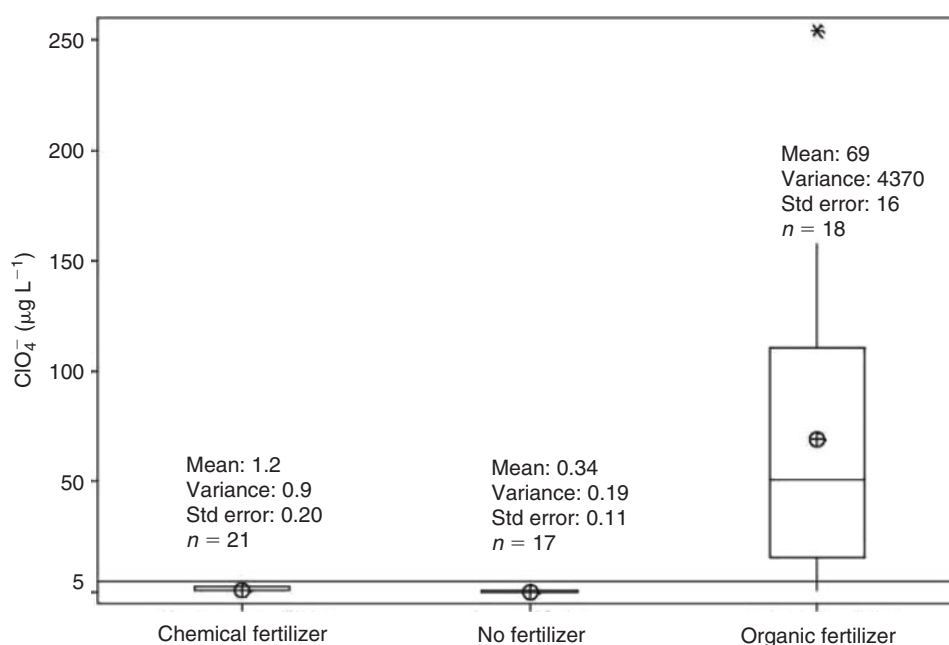


Fig. 3. Box plot of soil water data collected beneath maintained lawns.

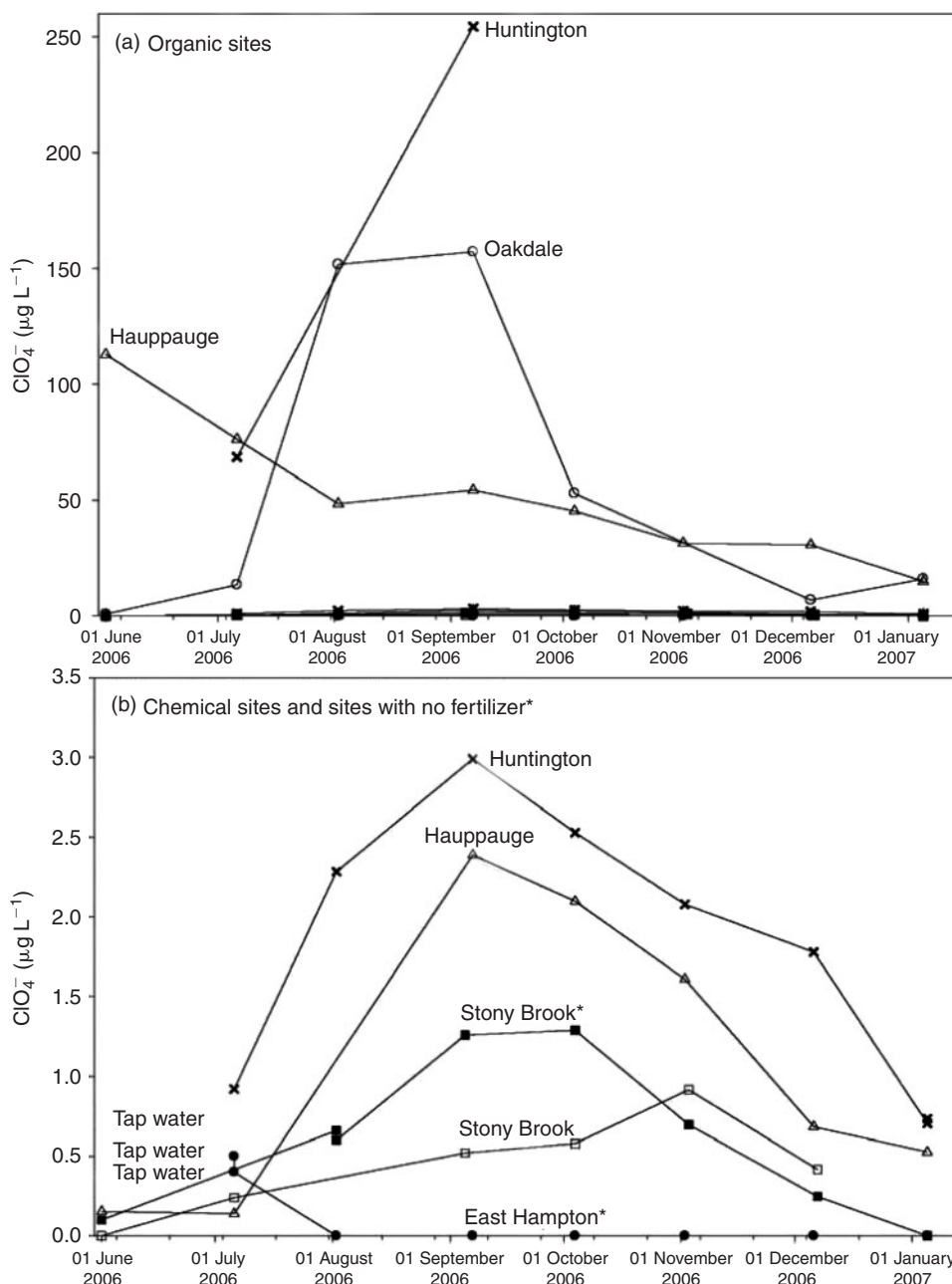


Fig. 4. Soil water perchlorate concentrations during the study period for (a) sites treated with organic fertilizer (the other sites are plotted but are not visible at this scaling); and (b) sites treated with chemical fertilizer or no fertilizer.

using a Lachat's QuickChem8500 Flow Injection Analysis System (Hach Company, Loveland, CO) with an uncertainty of 5% for N-NO_3^- and 10% for Cl determined by anonymous standards and duplicate analysis. Detection limits are $0.1 \text{ mg L}^{-1} \text{ N-NO}_3^-$ and $1 \text{ mg L}^{-1} \text{ Cl}$.

Results

Soil water concentrations and turfgrass content

Concentrations of perchlorate in soil water at 100 cm are highest beneath lawns treated with organic fertilizer, with an average concentration of 69 ± 16 (standard error of mean) $\mu\text{g ClO}_4^- \text{ L}^{-1}$ and a maximum concentration of $254 \mu\text{g L}^{-1}$ (Fig. 3). The average concentration of soil water collected beneath the sites treated with chemical fertilizer is $1.2 \pm 0.2 \mu\text{g L}^{-1}$. The average

concentration of soil water collected beneath the sites that were not fertilized is $0.34 \pm 0.44 \mu\text{g L}^{-1}$. Grass from the sites that were treated with organic fertilizer had the highest perchlorate concentrations of the treatment types, with an average concentration for oven-dried 'grass' of $3219 \pm 632 \mu\text{g ClO}_4^- \text{ kg}^{-1}$. The average concentration of grass from the sites treated with chemical fertilizer is $159 \pm 37 \mu\text{g kg}^{-1}$ and the average for the sites that were not fertilized is $120 \pm 39 \mu\text{g kg}^{-1}$. No clear correlation was observed between concentrations of ClO_4^- , Cl and N-NO_3^- in soil water collected at 100 cm or in grass. Tables A1, A2 and A3 in the Accessory publication list ClO_4^- , N-NO_3^- and Cl concentrations for the soil water and grass samples.

Concentrations of perchlorate vary in soil water collected beneath sites treated with organic fertilizer. The concentrations in samples from Huntington and Oakdale increase during the

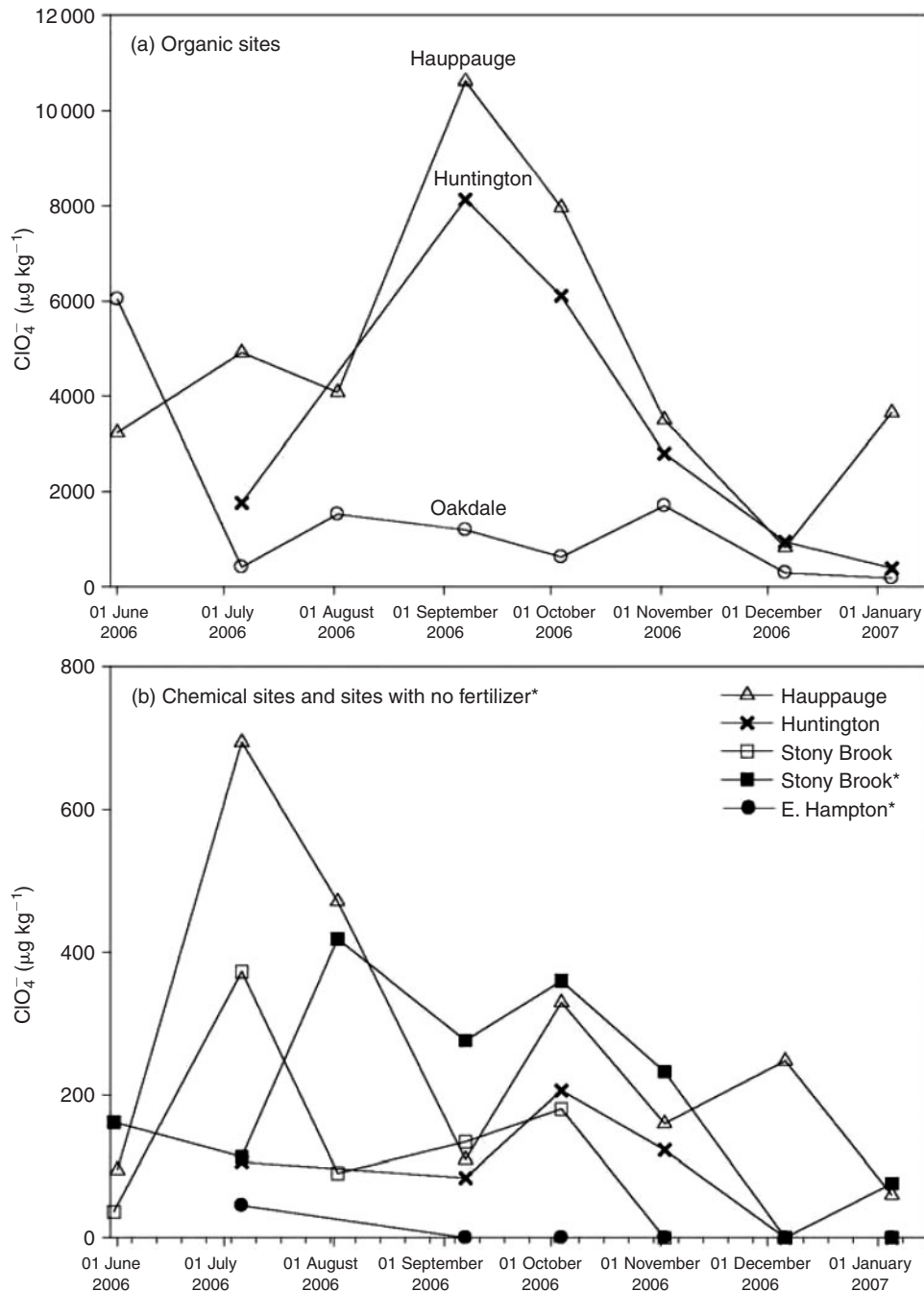


Fig. 5. Perchlorate concentration of oven-dried grass clippings for (a) sites treated with organic fertilizer; and (b) sites treated with chemical fertilizer or no fertilizer.

summer and peak in September. The samples from Hauppauge continually decrease through the summer and fall (Fig. 4a). The site at Hauppauge has more clay in the soil profile than do the other sites, which may slow water flow. The other sites have more sand. Estimated hydrologic conductivity based on soil types indicates that water at the surface should take less than the sampling period (30 days) to reach 100 cm for sites with predominately sand, whereas sites composed mostly of clay can take longer than the sampling period. Patterns of ClO_4^- concentrations of soil water were similar for the sites treated with chemical fertilizer and those not fertilized. They increased during the summer and peaked in September, then decreased through the fall (Fig. 4b),

much like the sites treated with organic fertilizer at Huntington and Oakdale. Soil water concentrations at East Hampton were routinely below detection. There is no apparent correlation between soil water concentration and precipitation volume (data not shown).

In general, concentrations of perchlorate in the grass decrease in late fall for all sites (Fig. 5). This is likely because cool-season grasses stop growing in late fall. Peak concentrations occur later in the year at the sites treated with organic fertilizer (Fig. 5a) than those treated with chemical fertilizer (Fig. 5b), possibly owing to the different sources of perchlorate for the different treatment types. The variability of concentrations of perchlorate

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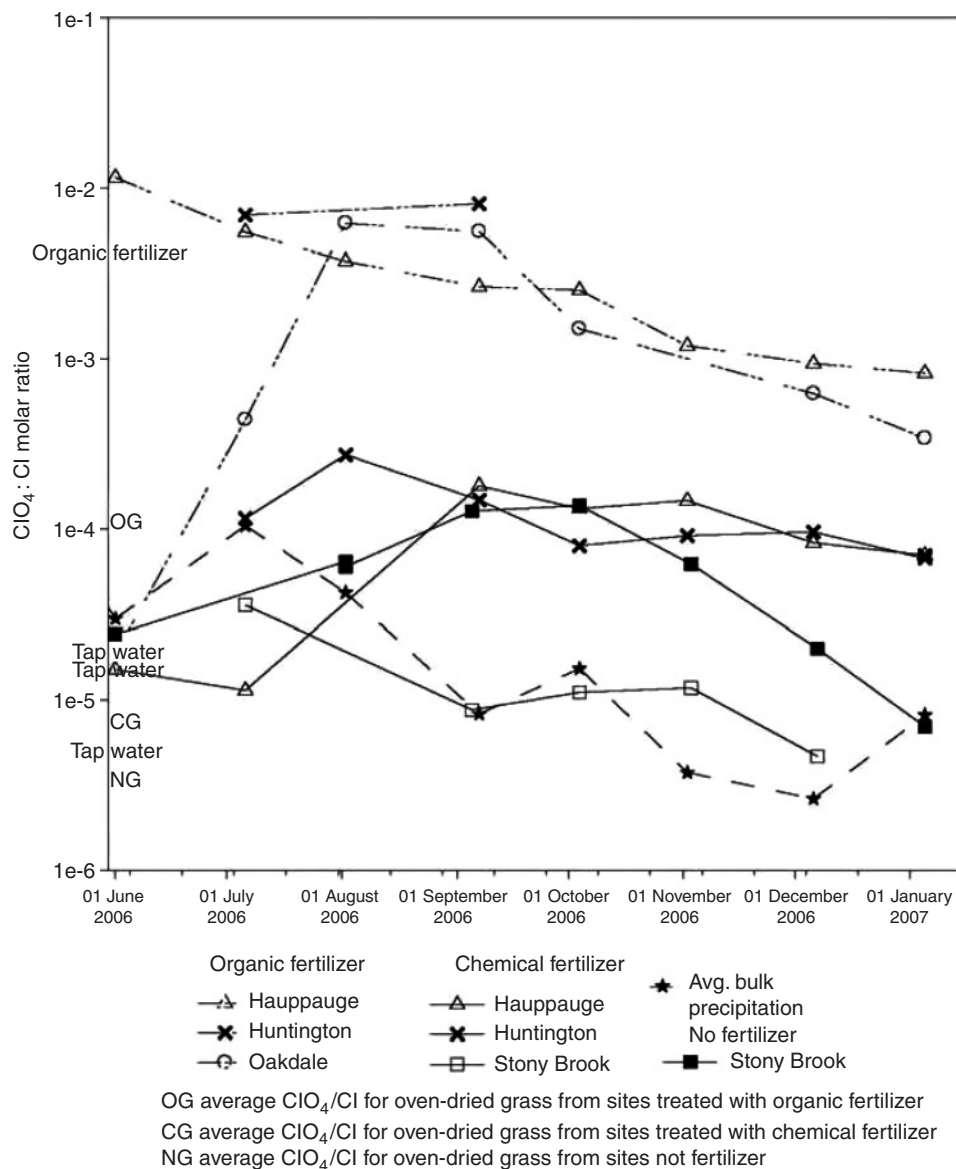


Fig. 6. ClO_4^- to Cl ratio of soil water samples. East Hampton, treated with no fertilizer, was not plotted because ClO_4^- was only measured in one sample. For reference tap water, organic fertilizer, the average value of bulk precipitation and the average values of oven-dried grass are plotted. The Cl content of the chemical fertilizer is unknown.

in grass between the sites may be due to variability in plant types and differences in the health of the ecosystems, neither of which were evaluated, as growth rates in grasses are dependent on the plant types, availability of water and soil temperature.

Perchlorate to chloride ratios might be expected to aid in understanding sources of perchlorate in the soil water. Any increase in the ClO_4^- to Cl ratio from bulk precipitation indicates another source of ClO_4^- besides precipitation because evaporation should not change this ratio. This ratio in soil water collected beneath sites that were fertilized with organic fertilizer is higher than that in bulk precipitation (average ratios shown on Fig. 6) and is similar to that in organic fertilizer (Fig. 6). Organic fertilizer used in the present study measured $9000 \mu\text{g ClO}_4^- \text{kg}^{-1}$. Soil water ClO_4^- to Cl ratios collected beneath sites that were fertilized with chemical fertilizer or not fertilized indicate another source of ClO_4^- besides bulk precipitation or tap water (Fig. 6). The ClO_4^- to Cl ratio in chemical fertilizer was not measured

but perchlorate concentrations ranged from not detected to $3 \mu\text{g ClO}_4^- \text{kg}^{-1}$.

The lawns were mowed from April to November with the mulched grass left on the lawns. The mulched grass may act as a reservoir for perchlorate. Grass takes a few weeks to decompose owing to its high lignin content. The breakdown increases the source of nutrients available for leaching or uptake by the live grass. Approximately 20–30% of the nitrogen and carbon in turfgrass clippings mineralises within 7 days after being cut, with decomposition rates depending on whether the clippings remain on or within the turf canopy, or whether they are transported to the soil surface.^[24] The ClO_4^- to Cl ratio of mulched grass is assumed to be that of live grass (Fig. 6). Perchlorate to chloride ratios of the organic-fertilized grasses are much lower than those of the organic fertilizer but higher than that of bulk precipitation. The ratios for the grasses that are fertilized with chemical fertilizer might have an unknown source of chloride,

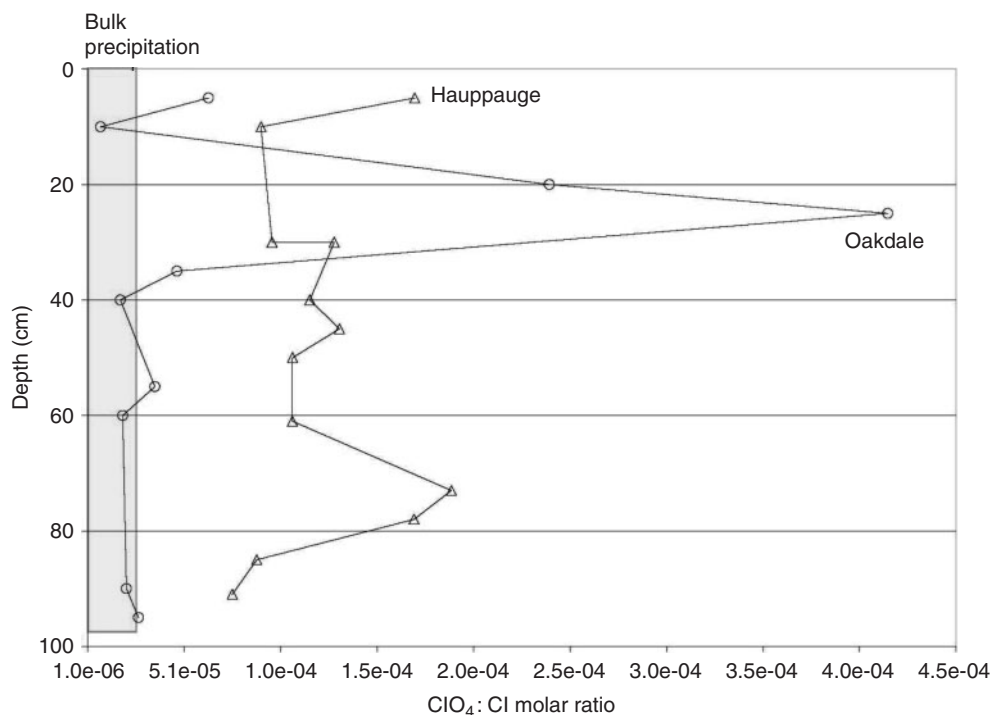


Fig. 7. ClO_4^- to Cl ratio of the two soil cores from sites treated with organic fertilizer. The range for bulk precipitation is plotted.

or there is some process that is reducing the ClO_4^- to Cl ratio in the grass.

Soil core data for organically fertilized sites

The analytical data, soil moisture, grain size analysis and porosity for the two soil cores are available in the Accessory publication (Table A4). The samples were collected in November 2006, 1 month after the sites had been fertilized. The abundance data for ClO_4^- , Cl, and N-NO_3^- in the core should reflect the amount of ClO_4^- , Cl, and N-NO_3^- that is in the soil moisture. This assumes that these anions are not adsorbed on the soil particles and will completely dissolve in the deionised water. They are abundances rather than concentrations because the ClO_4^- , Cl, and N-NO_3^- are not the concentrations in the soil moisture, but in the fixed amount of water that was added to a fixed amount of dry sample. Core samples with higher organic material or clay should be expected to have higher soil moisture and thus higher chloride abundance. Although there is not a simple correlation with chloride abundances and measured soil moisture, there is a general trend of increased chloride abundance with soil moisture. The ClO_4^- to Cl ratios are much lower than that of the organic fertilizer, which is $\sim 5 \times 10^{-3}$, but greater than that of bulk precipitation, which is 10^{-4} to 10^{-6} (Fig. 7).

Discussion

The concentrations of perchlorate in soil water and turfgrass at sites treated with chemical fertilizer and no fertilizer are much less than the concentrations at sites treated with organic fertilizer. The pattern of perchlorate in soil water and grass is similar at all sites. Initially, the turfgrass takes up perchlorate from an increase in the concentration of perchlorate in precipitation in July (associated with fireworks in the surrounding areas^[25]) or from the organic fertilizer applied in May. The grass is likely taking up

ClO_4^- via mass flow^[17] as the N-NO_3^- uptake pattern is not similar to ClO_4^- . Then, the mulched grass decomposes, providing an additional source of perchlorate to the lawn environment from May to November. Concentrations of perchlorate in the live grass generally follow the growing cycle of cool-season grasses but are also dependent on availability of perchlorate.^[15] What the grass does not take up eventually leaches into the unsaturated zone, as detected in the soil water at 100 cm, and through Long Island's sandy subsoil eventually to the groundwater. Thus, urban lawns fertilized with fertilizers high in perchlorate may adversely affect local groundwater quality.

Acknowledgements

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