L’oxydation, mais de manière maîtrisée - une thèse primée

17 juillet 2024 | Andri Bryner
Catégories: Eau potable | Eaux usées | Polluants | Organisation et personnel

La chimiste de l’environnement Joanna Houska a été récompensée pour sa thèse de doctorat par la Deutsche Wasserchemische Gesellschaft (Société allemande de Chimie de l’Eau). Joanna Houska a mené des recherches à l’Eawag et à l’EPFL, et a démontré tant théoriquement qu’expérimentalement comment le traitement oxydatif de l’eau avec de l’ozone ou du chlore peut être utilisé de manière plus efficace lorsqu’on connaît plus précisément les substances organiques dissoutes dans l’eau à traiter

«Ce travail est un jalon vers une meilleure compréhension du rôle de la matière organique dissoute dans le traitement oxydatif de l’eau», déclare Urs von Gunten, le directeur de thèse de Joanna Houska. Il est professeur à l’EPFL et dirige à l’Institut de recherche de l’eau Eawag le groupe Chimie de l’eau potable. Selon von Gunten, Joanna Houska possède «une énorme capacité à traiter des questions scientifiques complexes de manière théorique et expérimentale et à résoudre des problèmes avec une grande indépendance.» Dans ses expériences, mais aussi grâce à la modélisation, la chercheuse a produit et interprété de nouvelles données passionnantes qui peuvent être utilisées pour améliorer le traitement oxydatif de l’eau.

Inefficace et dangereux sans analyse précise

Quel est donc le problème? Lorsqu’on utilise du chlore ou de l’ozone pour la désinfection/l’oxydation de l’eau, le principal objectif est de neutraliser les germes pathogènes et les micropolluants. Mais la majeure partie des agents oxydants réagit avec les composants de la matrice de l’eau, tels que la matière organique dissoute. Cela réduit non seulement l’efficacité des procédés, mais peut aussi, dans le pire des cas, entraîner la formation de sous-produits toxiques. Il est donc important de savoir le plus précisément possible ce qui est dissous dans l’eau à traiter afin de rendre l’oxydation efficace. C’est là que la chercheuse Joanna Houska est intervenue, car jusqu’à présent, on utilisait principalement des paramètres globaux dérivés, par exemple, de l’absorption UV. Dans sa thèse, elle a donc tout d’abord développé des méthodes destinées à mesurer la concentration de composés pertinents. Elle les a ensuite caractérisés et a évalué s’ils pouvaient former des sous-produits problématiques, et dans quelle mesure. Enfin, elle a découvert, grâce aux isotopes de l’oxygène dans le peroxyde d’hydrogène, comment les précurseurs sensibles peuvent être distingués les uns des autres. Il est en outre intéressant de noter que Joanna Houska n’a pas limité sa recherche uniquement à la désinfection/l’oxydation de l’eau destinée à être utilisée comme eau potable. Bien plus, elle a également suivi, dans une station d’épuration, la formation de sous-produits de l’ozonation et leur dégradation ultérieure lors du traitement biologique secondaire des eaux usées épurées.

Pour la première fois en Suisse

Le Promotionspreis (en français: prix de thèse) dans le domaine de la chimie de l’eau est décerné depuis 1992 et est doté de 1500 euros. En 2024, il va pour la première fois à la Suisse. Joanna Houska sait déjà ce qu’elle fera de cet argent: «Mon vélo a besoin d’une mise à niveau urgente, donc l’argent du prix sera probablement investi là-dedans». Elle travaille désormais dans le département Environnement de Roche, dans le domaine des eaux usées et de la protection des eaux.

Photo de couverture: Joanna Houska lors de la remise du prix avec le Prof. Thomas Ternes au congrès annuel de la Wasserchemische Gesellschaft à Limburg (D) (Photo: Nina Hermes, BfG).

Publication originale

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disinfection or micropollutant abatement dur
        ing drinking water and wastewater treatment is accompanied by oxidation of m
        atrix components such as dissolved organic matter (DOM). To improve predicti
        ons of the efficiency of oxidation processes and the formation of oxidation
        products, methods to determine concentrations of oxidant-reactive phenolic,
        olefinic or amine-type DOM moieties are critical.<br />Here, a novel selecti
        ve oxidative titration approach is presented, which is based on reaction kin
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        etics of oxidation reactions towards certain DOM moieties. Phenolic moieties
         were determined by oxidative titration with ClO<sub>2</sub> and O<sub>3</su
        b> for five DOM isolates and two secondary wastewater effluent samples. The
        determined concentrations of phenolic moieties correlated with the electron-
        donating capacity (EDC) and the formation of inorganic ClO<sub>2</sub>-bypro
        ducts (HOCl, ClO<sub>2</sub><sup>?</sup>, ClO<sub>3</sub><sup>?</sup>).
        ClO<sub>2</sub>-byproduct yields from phenol and DOM isolates and changes du
        e to the application of molecular tagging for phenols revealed a better unde
        rstanding of oxidant-reactive structures within DOM.<br />Overall, oxidative
         titrations with ClO<sub>2</sub> and O<sub>3</sub> provide a novel and promi
        sing tool to quantify oxidant-reactive moieties in complex mixtures such as
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        f disinfection byproducts (DBPs) such as low molecular weight aldehydes and
        ketones from the reactions of ozone with dissolved organic matter (DOM). By
        applying a recently developed non-target workflow, 178 carbonous and nitroge
        nous carbonyl compounds were detected during bench-scale ozonation of two la
        ke waters and three secondary wastewater effluent samples and full-scale ozo
        nation of secondary treated wastewater effluent. An overlapping subset of ca
        rbonyl compounds (20%) was detected in all water types. Moreover, wastewater
         effluents showed a significantly higher fraction of <em>N</em>-containing c
        arbonyl compounds (30%) compared to lake water (17%). All carbonyl compounds
         can be classified in 5 main formation trends as a function of increasing sp
        ecific ozone doses. Formation trends upon ozonation and comparison of result
        s in presence and absence of the <sup>•</sup>OH radical scavenger DMSO in
        combination with kinetic and mechanistic information allowed to elucidate po
        tential carbonyl structures. A link between the detected carbonyl compounds
        and their precursors was established by ozonating six model compounds (pheno
        l, 4-ethylphenol, 4-methoxyphenol, sorbic acid, 3-buten-2-ol and acetylaceto
        ne). About one third of the detected carbonous carbonyl compounds detected i
        n real waters was also detected by ozonating model compounds.<br />Evaluatio
        n of the non-target analysis data revealed the identity of 15 carbonyl compo
        unds, including hydroxylated aldehydes and ketones (e.g. hydroxyacetone, con
        fidence level (CL) = 1), unsaturated dicarbonyls (e.g. acrolein, CL = 1;
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         2-butene-1,4-dial, CL = 1; 4-oxobut-2-enoic acid, CL = 2) and also a ni
        trogen-containing carbonyl compound (2-oxo-propanamide, CL =1).<br />Overall
        , this study shows the formation of versatile carbonous and nitrogenous carb
        onyl compounds upon ozonation involving ozone and <sup>•</sup>OH reactions
        . Carbonyl compounds wit...' (2196 chars) serialnumber => protected'0043-1354' (9
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of undesired byproducts from ozonation of dissolved organic matte
        r (DOM) such as aldehydes and ketones is currently hampered by limited knowl
        edge of their precursors and formation pathways. Here, the stable oxygen iso
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        ese byproducts was studied to determine if it can reveal this missing inform
        ation. A newly developed procedure, which quantitatively transforms H<sub>2<
        /sub>O<sub>2</sub> to O<sub>2</sub> for subsequent <sup>18</sup>O/<sup>16</s
        up>O ratio analysis, was used to determine the ?<sup>18</sup>O of H<sub>2</
        sub>O<sub>2</sub> generated from ozonated model compounds (olefins and pheno
        l, pH 3-8). A constant enrichment of <sup>18</sup>O in H<sub>2</sub>O<sub>2<
        /sub> with a ?<sup>18</sup>O value of ?59‰ implies that <sup>16</sup>O-
        <sup>16</sup>O bonds are cleaved preferentially in the intermediate Criegee
        ozonide, which is commonly formed from olefins. H<sub>2</sub>O<sub>2</sub> f
        rom the ozonation of acrylic acid and phenol at pH 7 resulted in lower <sup>
        18</sup>O enrichment (?<sup>18</sup>O = 47-49‰). For acrylic acid, enhanc
        ement of one of the two pathways followed by a carbonyl-H<sub>2</sub>O<sub>2
        </sub> equilibrium was responsible for the smaller ?<sup>18</sup>O of H<sub
        >2</sub>O<sub>2</sub>. During phenol ozonation at pH 7, various competing re
        actions leading to H<sub>2</sub>O<sub>2</sub> via an intermediate ozone addu
        ct are hypothesized to cause lower ?<sup>18</sup>O in H<sub>2</sub>O<sub>2<
        /sub>. These insights provide a first step toward supporting pH-dependent H<
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rbonyl compounds (aldehydes, ketones and ketoacids), a main class of organic
disinfection byproducts (DBPs). However, the detection of carbonyl compound
s in water and wastewater is challenged by multiple difficulties inherent to
their physicochemical properties. A non-target screening method involving t
he derivatisation of carbonyl compounds with p-toluenesulfonylhydra
zine (TSH) followed by their analysis using liquid chromatography coupled to
electrospray ionisation high-resolution mass spectrometry (LC-ESI-HRMS) and
an advanced non-target screening and data processing workflow was developed
. The workflow was applied to investigate the formation of carbonyl compound
s during ozonation of different water types including lake water, aqueous so
lutions containing Suwannee River Fulvic acid (SRFA), and wastewater. A high
er sensitivity for most target carbonyl compounds was achieved compared to p
revious derivatisation methods. Moreover, the method allowed the identificat
ion of known and unknown carbonyl compounds. 8 out of 17 target carbonyl com
pounds were consistently detected above limits of quantification (LOQs) in m
ost ozonated samples. Generally, the concentrations of the 8 detected target
compounds decreased in the order: formaldehyde > acetaldehyde > glyox
ylic acid > pyruvic acid > glutaraldehyde > 2,3-butanedione > gl
yoxal > 1-acetyl-1-cyclohexene. The DOC concentration-normalised formatio
n of carbonyl compounds during ozonation was higher in wastewater and SRFA-c
ontaining water than in lake water. The specific ozone doses and the type of
the dissolved organic matter (DOM) played a predominant role for the extent
of formation of carbonyl compounds. Five formation trends were distinguishe
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_versionedUid => protected30345 (integer)modified pid => protected124 (integer) Houska, J.; Salhi, E.; Walpen, N.; von Gunten, U. (2021) Oxidant-reactive carbonous moieties in dissolved organic matter: selective quantification by oxidative titration using chlorine dioxide and ozone, *Water Research*, 207, 117790 (11 pp.), doi:10.1016/j.watres.2021.117790, Institutional Repository

Houska, J.; Manasfi, T.; Gebhardt, I.; von Gunten, U. (2023) Ozonation of lake water and wastewater: identification of carbonous and nitrogenous carbonyl-containing oxidation byproducts by non-target screening, *Water Research*, 232, 119484 (17 pp.), doi:10.1016/j.watres.2022.119484, Institutional Repository

Houska, J.; Stocco, L.; Hofstetter, T. B.; Gunten, U. von (2023) Hydrogen peroxide formation during ozonation of olefins and phenol: mechanistic insights from oxygen isotope signatures,

Environmental Science and Technology, 57, 18950-18959, doi:10.1021/acs.est.3c00788, Institutional Repository

Manasfi, T.; Houska, J.; Gebhardt, I.; von Gunten, U. (2023) Formation of carbonyl compounds during ozonation of lake water and wastewater: development of a non-target screening method and quantification of target compounds, *Water Research*, 237, 119751 (14 pp.), doi:10.1016/j.watres.2023.119751, Institutional Repository

Links

Prix de thèse

Groupe de recherche Eawag sur la chimie de l'eau potable

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