



Prix Otto Jaag pour la protection des eaux 2023 et médaille EPF pour Charlotte Bopp

21 novembre 2023 | Claudia Carle
Catégories: Organisation et personnel

Charlotte Bopp, chercheuse en sciences de l'environnement, est doublement récompensée pour sa thèse: elle a reçu le 18 novembre le prix Otto Jaag pour la protection des eaux lors de la Journée de l'EPF et recevra également la médaille EPF en janvier. Son travail fournit une importante contribution à la compréhension de la biodégradation des polluants organiques dans l'environnement.

Avec le prix Otto Jaag pour la protection des eaux, l'EPF Zurich récompense l'excellence des mémoires de master et des thèses de doctorat dans le domaine de la protection des eaux et des sciences aquatiques. Cette distinction a été remise à Charlotte Bopp lors de la Journée de l'EPF le 18 novembre pour sa thèse intitulée «The role of oxygen uncoupling by Rieske non-heme iron dioxygenases in the biodegradation of aromatic contaminants». La médaille EPF attribuée par l'EPF Zurich aux mémoires de master et aux thèses de doctorat d'excellence lui sera également décernée en janvier.

Oxydation inefficace des polluants

En tant que doctorante à l'Institut de recherche sur l'eau Eawag au département Chimie de l'environnement, Charlotte Bopp a étudié de près la biodégradation de polluants organiques difficilement biodégradables. Si de tels composés aromatiques, présents par exemple dans les pesticides, les médicaments ou les explosifs, parviennent dans le sol et les cours d'eaux, les micro-organismes peuvent les oxyder et les dégrader grâce à un groupe d'enzymes appelées «oxygénases de Rieske». Dans son travail, la chercheuse s'est concentrée sur le sous-groupe d'enzymes dégradant les explosifs et a cherché à déterminer leur niveau d'efficacité. Ses résultats ne donnent pas

O_2 catalyzed by Rieske dioxygenases are frequent initial steps of biodegradation in natural and engineered environments. Many of these non-heme ferrous iron enzymes are known to be involved in contaminant metabolism, but the understanding of enzyme-substrate interactions that lead to successful biodegradation is still elusive. Here, we studied the mechanisms of O_2 activation and substrate hydroxylation of two nitroarene dioxygenases to evaluate enzyme- and substrate-specific factors that determine the efficiency of oxygenated product formation. Experiments in enzyme assays of 2-nitrotoluene dioxygenase (2NTDO) and nitrobenzene dioxygenase (NBDO) with methyl-, fluoro-, chloro-, and hydroxy-substituted nitroaromatic substrates reveal that typically 20-100% of the enzyme's activity involves unproductive paths of O_2 activation with generation of reactive oxygen species through so-called O_2 uncoupling. The ^{18}O and ^{13}C kinetic isotope effects of O_2 activation and nitroaromatic substrate hydroxylation, respectively, suggest that O_2 uncoupling occurs after generation of Fe^{III}-(hydro)peroxo species in the catalytic cycle. While 2NTDO hydroxylates *ortho*-substituted nitroaromatic substrates more efficiently, NBDO favors *meta*-substituted, presumably due to distinct active site residues of the two enzymes. Our data implies, however, that the O_2 uncoupling and hydroxylation activity cannot be assessed from simple structure-reactivity relationships. By quantifying O_2 uncoupling by Rieske dioxygenases, our work provides a mechanistic link between contaminant biodegradation, the generation of reactive oxygen species, and possible adaptation strategies of microorganisms to the exposure of new contaminants.' (1924 chars) serialnumber => protected" (0 chars) doi => protected'10.1021/acsenvironau.2c00023' (28 chars) uid => protected25093 (integer) _localizedUid => protected25093 (integer)modified _languageUid => protectedNULL _versionedUid => protected25093 (integer)modified pid => protected124 (integer) 1 => Snowflake\Publications\Domain\Model\Publicationprototypepersistent entity (uid=24911, pid=124) originalId => protected24911 (integer) authors => protected'Pati, S. G.; Bopp, C. E.; Kohler, H.-P. E.; Hofstetter, T. B.' (101 chars) title => protected'Substrate-specific coupling of O_2 activation to hydroxylations of aromatic compounds by rieske non-heme iron dioxygenases' (132 chars) journal => protected'ACS Catalysis' (13 chars) year => protected2022 (integer) volume => protected12 (integer) issue => protected'11' (2 chars) startpage => protected'6444' (4 chars) otherpage => protected'6456' (4 chars) categories => protected'non-heme ferrous iron oxygenases; nitrobenzene dioxygenase; biocatalysis; O_2 uncoupling; isotope effects; xenobiotics' (117 chars) description => protected'Rieske dioxygenases catalyze the initial steps in the hydroxylation of aromatic compounds and are critical for the metabolism of xenobiotic substances. Because substrates do not bind to the mononuclear non-heme Fe^{II} center, elementary steps leading to O_2 activation and substrate hydroxylation are difficult to delineate, thus making it challenging to rationalize divergent observations on enzyme mechanisms, reactivity, and substrate specificity. Here, we show for nitrobenzene dioxygenase, a Rieske dioxygenase capable of transforming nitroarenes to nitrite and substituted catechols, that unproductive O_2 activation with the release of the unreacte

d substrate and reactive oxygen species represents an important path in the catalytic cycle. Through correlation of O_2 uncoupling for a series of substituted nitroaromatic compounds with ^{18}O and ^{13}C kinetic isotope effects of dissolved O_2 and aromatic substrates, respectively, we show that O_2 uncoupling occurs after the rate-limiting formation of Fe^{III}-(hydro)peroxo species from which substrates are hydroxylated. Substituent effects on the extent of O_2 uncoupling suggest that the positioning of the substrate in the active site rather than the susceptibility of the substrate for attack by electrophilic oxygen species is responsible for unproductive O_2 uncoupling. The proposed catalytic cycle provides a mechanistic basis for assessing the very different efficiencies of substrate hydroxylation vs unproductive O_2 activation and generation of reactive oxygen species in reactions catalyzed by Rieske dioxygenases.' (1703 chars) serialnumber => protected'2155-5435' (9 chars) doi => protected'10.1021/acscatal.2c00383' (24 chars) uid => protected24911 (integer) _localizedUid => protected24911 (integer)modified _languageUid => protectedNULL _versionedUid => protected24911 (integer)modified pid => protected124 (integer) Bopp, C. E.; Bernet, N. M.; Kohler, H.-P. E.; Hofstetter, T. B. (2022) Elucidating the role of O_2 uncoupling in the oxidative biodegradation of organic contaminants by Rieske non-heme iron dioxygenases, *ACS Environmental Au*, 2(5), 428-440, doi:10.1021/acsenvironau.2c00023, [Institutional Repository](#)

Pati, S. G.; Bopp, C. E.; Kohler, H.-P. E.; Hofstetter, T. B. (2022) Substrate-specific coupling of O_2 activation to hydroxylations of aromatic compounds by rieske non-heme iron dioxygenases, *ACS Catalysis*, 12(11), 6444-6456, doi:10.1021/acscatal.2c00383, [Institutional Repository](#)

Links

Prix Otto Jaag pour la protection des eaux

Page du projet "Enzyme Mechanisms and Kinetics of Organic Contaminant Oxygenation"

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