Aromatic tertiary amines are one of the most important classes of organic compounds in organic chemistry and drug discovery. It is difficult to efficiently construct tertiary amines from primary amines via classical nucleophilic substitution due to consecutive overalkylation. In this paper, we have developed a radical tandem C-N coupling strategy to efficiently construct aromatic tertiary amines from commercially available carboxylic acids and nitroarenes. A variety of aromatic tertiary amines can be furnished in good yields (up to 98%) with excellent functional group compatibility under mild reaction conditions. The use of two different carboxylic acids also allows for the concise synthesis of nonsymmetric aromatic tertiary amines in satisfactory yields. Mechanistic studies suggest the intermediacy of the arylamine–(TPP)Fe(III) species and might provide a possible evidence for an S_{H2} (bimolecular homolytic substitution) pathway in the critical C-N bond formation step.
Aromatic tertiary amines, especially bearing benzylic units, are versatile building blocks in organic synthesis, as they contain differentiable C(sp³)-H, C-N bond, and C(sp²)-H bonds. The biological activity of aromatic tertiary amines and their ability to influence natural neurotransmitter transmission routes enable them highly effective for drug discovery, in areas that range from the treatment of bone deterioration (such as a fracture) to cytopenia (such as cancer) (Fig. 1a). Consequently, the development of synthetic methods to forge C-N bonds for the synthesis of tertiary amines has gained considerable attention in recent decades. Classical synthetic methods to access these tertiary amines are mainly derived from the reductive amination or nucleophilic substitution from aromatic amines that are produced industrially from nitroarenes. However, the nucleophilic substitution of aromatic amines with benzylic halides may result in overalkylation side reactions to some extent, and despite being an efficient process, reductive amination requires the use of reactive aldehydes and reductants.

Nitro(hetero)arenes are readily available and commercially cheap feedstock reagents. They have recently been used as amination reagents with the reduction in situ, efficiently avoiding the hydrogenation step and thus improving step-economy and reducing cost. The direct use of nitrobenzene as potential amines has gained highly active research interests. Besides the classical ionic-type reductive amination with nitroarenes, the emerging radical reductive amination of nitroarenes with alkenes and organic halides has been successfully developed, however only practical for the construction of secondary amines (Fig. 1b). To the best of our knowledge, the direct use of nitroarenes for the construction of tertiary amines is still underdeveloped. Its success would represent a significant step forward to simplify tertiary amines synthesis from bench-stable and commercially cheap nitroarenes.

Herein, we develop a highly efficient catalytic strategy for decarboxylative C-N coupling using both commercially cheap carboxylic acids and nitroarenes, delivering a wide range of aromatic tertiary amines in 35–98% yields (Fig. 1c). Moreover, the use of two different carboxylic acids can allow for the concise synthesis of non-symmetric tertiary amines in synthetically acceptable yields.

**Results**

**Reaction optimization.** To initiate our study, we selected phenylacetic acid (1) and nitrobenzene (2) as model substrates with which to explore the optimized reaction conditions (Table 1). After a series of conditional screening, the standard conditions include 10 mol% FeI₂, 1 mol% 4CzIPN, 10 mol% ligand (L₁), and reductant (EtO)₃SiH by means of irradiation of blue LEDs at 65 °C (entry 1). Under the standard conditions, the desired amine (4) can be obtained in 91% isolated yield. Notably, the classical SN₂ nucleophilic substitution reaction of aromatic amines and benzylic halides is challenging to deliver tertiary amine (4) because of the competing overalkylation, instead of the formation of a quaternary ammonium salt. Under the optimized conditions, the desired amine (4) can be obtained in 91% isolated yield. Notably, the classical SN₂ nucleophilic substitution reaction of aromatic amines and benzylic halides is challenging to deliver tertiary amine (4) because of the competing overalkylation, instead of the formation of a quaternary ammonium salt. Under the optimized conditions, the use of other ligands (L₁ to L₅) can decrease the yield (entries 2–5, Table 1). It is found that FeI₂ is an important factor for the...
success as replacing with other iron-based catalysts resulted in a reduction of yields (entries 6, 7 and 10). We envisioned that ligands might affect the ligand-metal-charge-transfer (LMCT) process in the excited state of iron-complex. Also, we found that other silanes in place of (EtO)₃SiH cannot further increase the absorption peak at 412 nm, while their complex of FeI₂ and absorption peaks at 292 nm and 363 nm. The ligand (ligand) has been reported by others. Also crucial but can improve reaction efficiency. This phenomenon has also been reported by others. We also envisioned that the photocatalytic cycle could increase the concentration of benzyl radical (from decarboxylation of the carboxyl radical). The control experiment confirmed that the decarboxylative C-N coupling requires light irradiation since no reaction occurred in the dark otherwise under optimized conditions (entry 11). In addition, it is found that ligand (L1) has a great influence on the reaction yield. As a consequence, we measured the UV-Vis spectra of FeCl₃, ligand (L1), and their complexes. It is found that FeCl₃ has two absorption peaks at 292 nm and 363 nm. The ligand (L1) has one absorption peak at 412 nm, while their complex of FeL₂ and L₁ has a new absorption peak at 513 nm (Supplementary Fig. 7). We speculated that the addition of ligand can not only affect the LMCT process but also may play a key role in the subsequent reduction of N-based intermediates.

**Substrate scope.** With the optimal conditions in hand, we explored the substrate’s scope of carboxylic acids and nitro compounds in an orthogonal way. The representative examples are shown in Fig. 2. In general, this decarboxylative C-N coupling protocol holds satisfactory functional group compatibility. A wide range of carboxylic acids and nitroarenes can proceed with this decarboxylative C-N coupling to construct tertiary amines (4–48) in moderate to excellent yields (45 examples, up to 98%). It was found that the substituents of nitroarenes on ortho-, meta-, and para-position on the phenyl rings can all occur in this transformation. More importantly, the stERICALLY hindered methyl (10), n-butyl (11), and i-propyl (12) groups at the ortho-position of nitroarenes have little influence on the reaction efficiency. This indicates that steric hindrance hardly decreased the reactivity. A great number of versatile functional groups, such as ether (16, 23, 24), sulfide (14, 15, 18), hydroxyl (25–28), bromo (29), chloro (30), fluoro (31, 34), ester (37–39), ketone (40), amide (41), heterocycle (42–43, 47–48), boronic acid ester (45), and alkynyl (46) remain intact during decarboxylative C-N couplings. These useful functional groups enable this protocol promising for the synthesis of highly functionalized tertiary amines. Another interesting feature of this decarboxylative tertiary amine synthesis is that it has excellent selectivity among several nucleophilic functional groups. For example, the electron-rich NH-free indole-based nitro compound can be applied for this decarboxylative C-N coupling (42), which readily undergoes nucleophilic addition and substitution. The tertiary amines units that are generally incompatible in traditional nucleophilic substitution reactions are also suitable for this strategy. In addition, nitroarenes bearing active C-H bonds also tolerated the reaction conditions well (25–28, 36, 39–41).

Besides the good functional group tolerance concerning nitroarenes, it was also found that a set of functionalized carboxylic acids are competent coupling partners as shown in Fig. 2. Both the electron-rich and -poor functional groups on the phenyl rings (4–48) can undergo this decarboxylative C-N coupling process,
Fig. 2 Scope of the carboxylic acids and the nitro compounds. Reaction conditions: 4CzIPN (1 mol%), FeI₂ (10 mol%), L₁ (10 mol%), nitrobenzene (0.2 mmol), carboxylic acid (0.6 mmol), 2,6-lutidine (3.0 equiv.), (EtO)₃SiH (2.0 equiv.), MeCN (2 mL), blue LEDs, 65 °C, 24 h. Gram-scale test with 0.2 mol% photocatalyst. Standard conditions for gram-scale experiment: PC 3 (0.2 mol%), L₁ (10 mol%), FeI₂ (10 mol%), phenylacetic acid 1 (15 mmol), nitrobenzene 2 (5 mmol), 2,6-lutidine (15 mmol), (EtO)₃SiH (10 mmol), MeCN (25 mL), blue LEDs, 65 °C, 72 h. Reaction time = 45 h, 25 °C.
Fig. 3 Representative examples for three-component coupling reactions. Isolated yield (main products: 49-63; by-products: 49a-63a, 49b-63b) after chromatography were shown in turn. Reaction conditions: 4CzIPN (1 mol%), FeI2 (10 mol%), L1 (10 mol%), carboxylic acid A (0.3 mmol), carboxylic acid B (0.3 mmol), nitroarenes (0.2 mmol), 2,6-lutidine (3.0 equiv.), (EtO)3SiH (2.0 equiv.), MeCN (2 mL), Blue LEDs, 65 °C, 24 h. aReaction time = 32 h.
where the electron-rich groups usually give rise to the desired products in relatively better yields. We postulated that the electron-withdrawing groups on the arenes would decrease the nucleophilic features of the resulting benzylic radical, which is a key intermediate after decarboxylation. A wide range of substituted phenylacetic acid derivatives have been employed in this protocol and they can undergo this decarboxylative C-N coupling smoothly, affording the desired products (4–48) in 46–98% yields. Furthermore, with modified standard conditions using only 0.2 mol% 4CzIPN, a scaled-up experiment of 5 mmol can be conducted smoothly to afford the desired product (4) in 75% yield. However, due to the shorter lifetime of alkyl radicals, it is still difficult for common alkyl carboxylic acids at present.

Subsequently, two kinds of carboxylic acids have been employed to investigate a three-component reaction, which is able to construct non-symmetric aromatic tertiary amines (Fig. 3). Considering the differences in decarboxylation rates and generated radical stability of aromatic acetic acids with different substituents, we subjected two different carboxylic acids to this protocol, successfully affording non-symmetric aromatic tertiary amines (15 examples) in 35–59% yield. It was found that secondary carboxylic acids were also tolerated. They can give rise to the desired products (55–63) in acceptable results, indicating that the steric hindrance of the aromatic benzyl group hardly affected the success of the radical decarboxylative C-N coupling reaction. Besides the cross-coupling products of two aryl acetic acids (64–67), the coupling with alkyl acetic acids (68–73) also provided a diverse set of products (74–75).

Fig. 4 Synthetic application of derivatization of aromatic tertiary amines. [M] = Metal reagent. See Supplementary Information for detailed experimental conditions.
**Fig. 5** Mechanism study. **a** Controlled experiment. **b** Effects of various iron catalysts, ligand L1, and additive Nal on reaction and the study of S_{3}2. **c** Crossover experiment between 80 and 81. **d** The three-component cross-coupling. **e** Plausible mechanism.
acids, a small amount of homocoupling by-products (49a–63a, 49b–63b) were detected (Supplementary Fig. 3).

**Synthetic application.** The versatile transformations of aromatic tertiary amines as building blocks in the field of organic synthesis are explored (Fig. 4). Three different amides (64–66) and triazine (67) were obtained through selective C-N bond functionalization. Similarly, the amino group, as one of the main groups regulating the electrical properties of aromatic rings, can significantly affect the sp² C-H bonds of para positions. Several new valuable compounds (68, 70, 72, 73) have been obtained via controllable para-C-H functionalization of aromatic tertiary amines. Also, the α-C-H bonds adjacent to nitrogen atoms can be easily activated, furnishing products (69, 71, 75) in moderate yields. The cyclic aromatic tertiary amines (74) can also be obtained by aromatic ring dehalogenation coupling from product 34. The success of these derivatized products suggests the potential application of this protocol in organic synthesis.

**Mechanistic studies.** In order to explore the mechanism of the reaction, the following experiments were carried out. First, under the standard conditions, when 2 equiv. of TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl radical) was added into the reaction mixture, the reaction was completely inhibited and the corresponding methylpiperidine-1-oxyl radical was added into the reaction mixture (Supplementary Fig. 4), suggesting the success of the mechanism experiments showed that nitrobenzene (2) cannot be reduced to aniline under experimental conditions. On the other hand, when aniline was employed to react with benzene (4), the reaction was completely inhibited and the corresponding nitrobenzene (77) was added into the reaction mixture to replace nitrobenzene, the target product (4) was obtained in 45% yields. Accordingly, we hypothesized that nitrosobenzene (77) may be one important intermediate of this reaction (Fig. 5a).

Interestingly, when Fe(TPP)(III)Cl was used as the catalyst, no decarboxylative C-N coupling occurred (Fig. 5b). However, when 50 mol% amount of NaI was added to the above mixture, the desired reaction can readily proceed to afford the target product (4) in 42% yield. This may imply that the presence of iodide anion is important for such transformation. Accordingly, corresponding Fe-amine complex (80) and Fe-carboxylate complex (81) were prepared, respectively. Under the irradiation of blue light, the desired C-N bonds coupling between Fe-amine complex (80) and Fe-carboxylate complex (81) smoothly proceed to afford the product (4) in 46% yield (Fig. 5c). On the other hand, the byproduct, 1,2-diphenylethane (82) was formed during this process, suggesting the success of the decarboxylative process to generate benzyl radical from Fe-carboxylate complex (81). It is worth noting that blue light and NaI are crucial conditions for this reaction. Finally, the three-component reaction of nitrobenzene (2), the carboxylic acid (83) and Fe-carboxylate complex (81) can successfully give rise to the desired cross-coupling product (84) in 10% yield, which again confirms the Fe-carboxylate complex (81) would be a potential intermediate of the reaction (Fig. 5d).

According to the above mechanism experiments, a plausible mechanism was proposed and is shown in Fig. 5e. Nitro compounds are able to be reduced by (TPP)Fe(II) catalyst with (EtO)₃SiH to form Fe-based complexes (85)³⁴. The alkyl radical, which is generated via either the visible-light-mediated single-electron oxidation of carboxylic anion or light-mediated LMCT process of Fe-carboxylate (87), then attacks the 85 or 77 (Fig. 5a) to form the Fe(III) complex (86)³⁴,³⁹. Subsequently, it can undergo anion exchange with carboxylic anion to produce the hydroxylamine compound (88) and regenerate the Fe-species (87). Alternatively, the direct radical addition of alkyl radical to nitrosobenzene and subsequent recombination with (TPP)Fe(II) to produce 86 is also likely. The excited complex of carboxylic acid and iron(III) (87) proceeds through homolysis to form alkyl radical and iron catalyst (TPP)Fe(II) through a ligand-to-metal charge transfer (LMCT) pathway⁴⁹–⁵³. Simultaneously, in the light of Baran’s pioneering work, the resulting hydroxylamine compound (88) is able to react with (TPP)Fe(II) species to generate secondary intermediate (90) and (TPP)Fe(III) (See Supplementary Information)⁵³. In the presence of iodide anion, the resulting (TPP)Fe(III) species and intermediate (90) may recombine to form the Fe-based complex (91). With the irradiation of blue LEDs, the homolytic SₓP₂⁶⁰–⁶⁶ process can occur between 91 and alkyl radical, resulting in the formation of tertiary amines and the regeneration of (TPP)Fe(II). The by-products of the reaction might also confirm this mechanism (Supplementary Fig. 5).

**Discussion**

In summary, we have developed a method to construct two C-N bonds at once enabled by the catalysis of (TPP)Fe-complex with light irradiation, efficiently constructing a variety of synthetically valuable tertiary amines (up to 98% yields). A wide range of bench-stable carboxylic acids and nitroarenes are competent coupling partners in this decarboxylative C-N coupling. The use of two different carboxylic acids allows for the concise synthesis of nonsymmetric tertiary amines in synthetically useful yields. It represents a complementary manner to construct aromatic tertiary amines and can avoid the overallkylation limitation of the classical nucleophilic substitution process of anilines. The mechanistic studies suggest an SₓP₂ pathway, and thus hold advances in the selectivity among different nucleophilic functional groups. The detailed mechanistic study and exploration of this meaningful bonding-formation manner is ongoing in our group.

**Methods**

**General procedure for amidation.** 4-CzIPN (1.58 mg, 1 mol%), L1 (12.3 mg, 10 mol%), FeL₂ (6.2 mg, 10 mol%) were placed in an 8 mL transparent vial equipped with a stirring bar. This vial was carried into the glovebox, which is equipped with nitrogen. Then aromatic acid (0.6 mmol), nitroarenes (0.2 mmol), MeCN (1.0 mL), 2.6-lutidine (70 μL, 0.6 mmol) and (EtO)₃SiH (40 μL, 0.4 mmol), were added in sequence under N₂ atmosphere. The reaction mixture was stirred under the irradiation of blue LEDs (distance app. 5.0 cm from the bulb) at 65 °C for 24 h. When the reaction finished, the mixture was quenched with water and it was extracted with ethyl acetate (3 × 10 mL). The organic layers were combined to give the crude product, which was purified by flash column chromatography on silica gel (petroleum ether: ethyl acetate).

**Data availability**

We declare that all other data supporting the findings of this study are available within the article and Supplementary Information files.

Received: 30 December 2021; Accepted: 18 April 2022; Published online: 04 May 2022

**References**

1. Liu, S., Zhao, Z. & Wang, Y. Construction of N-heterocycles through cyclization of tertiary amines. *Chem. Eur. J.* 25, 2423–2441 (2019).
2. Balsam, P. et al. Antazoline for termination of atrial fibrillation during the procedure of pulmonary veins isolation. *Adv. Med. Sci.* 60, 231–235 (2015).
3. Kayser-Brickera, K. J. et al. Non-peptidic substrate-mimetic inhibitors of Akt as potential anti-cancer agents. *Bioorg. Med. Chem. Lett.* 17, 1764–1771 (2007).

4. Ruiz-Castillo, P. & Buchwald, S. L. Applications of palladium-catalyzed C-N cross-coupling reactions. *Chem. Rev.* 116, 12564–12649 (2016).

5. Roy, A. et al. Practical and regioselective amination of arenes using alkyl amines. *Nat. Chem.* 11, 426–433 (2019).

6. S. U. D., Julia, F., Luridiana, A., Douglas, J. J. & Leonori, D. A photochemical hydrodynamogenic strategy for amine synthesis. *Nat. Chem.* 358, 75–81 (2020).

7. Forero-Cortés, P. A. & Haydl, A. M. The 25th anniversary of the Buchwald–Hartwig amination: development, applications, and outlook. *Org. Process Res. Dev.* 23, 1478–1483 (2019).

8. Cheung, C. W., Leendert Ploeger, M. & Hu, X. Amide synthesis via nickel-catalyzed hydroamination of alkenes. *Angew. Chem. Int. Ed.* 58, 7532–7537 (2019).

9. Liu, Y. et al. Light-promoted C-N coupling of aryl halides with nitroarenes. *Angew. Chem. Int. Ed.* 60, 5230–5234 (2021).

10. Zhang, M., Yuan, X.-A., Zhu, C. & Xie, J. Deoxygenative deuteriation of carboxylic acids with D2O. *Angew. Chem. Int. Ed.* 58, 312–316 (2019).

11. Mao, R., Balon, J. & Hu, X. Cross-coupling of alkyl redox-active esters with benzenophenone imines: tandem photoredox and copper catalysis. *Angew. Chem. Int. Ed.* 57, 9501–9504 (2018).

12. Zhang, W., Wurz, R. P., Pettit, J. C. & Fu, G. C. Photoinduced, copper-catalyzed decarbonylation of conjugated C-N coupling to generate protected amines: an alternative to the curtius rearrangement. *J. Am. Chem. Soc.* 139, 12153–12156 (2017).

13. Priester, K. C., Rankic, D. A. & MacMillan, D. W. Visible light photoredox catalysis with transition metal complexes: applications in organic synthesis. *Chem. Rev.* 113, 5322–5363 (2013).

14. Skibi, K. L., Blum, T. R. & Yoon, J. P. T. P. Dual catalysis strategies in photochemical synthesis. *Chem. Rev.* 116, 10035–10074 (2016).

15. Yu, X.-Y., Chen, J.-R. & Xiao, W.-J. Visible light-driven radical-mediated C–C bond cleavage/function alization in organic synthesis. *Chem. Rev.* 121, 506–561 (2021).

16. Wang, C.-S., Dineux, P. H. & Soule, J.-F. Photoredox catalysis for building C–C bonds from C(sp3)–H bonds. *Chem. Rev.* 118, 7532–7585 (2018).

17. Liu, W., Li, J., Huang, C.-Y. & Li, C.-J. Aromatic chemistry in the excited state: facilitating metal-free substitutions and cross-couplings. *Angew. Chem. Int. Ed.* 132, 1802–1812 (2019).

18. Abdareykov, B., Bhattacharyya, A. & Reiser, O. Visible-light-induced homolysis of earth-abundant metal-substrate complexes: a complementary activation strategy in photoredox catalysis. *Angew. Chem. Int. Ed.* 60, 21100–21115 (2021).

19. Feng, G., Wang, X. & Jin, J. Decarbonylation C-C and C-N bond formation by ligand-activated iron photocatalysis. *Eur. J. Org. Chem.* 2019, 6728–6732 (2019).

20. Jin, Y. et al. Convenient C(sp3)–H bond functionalisation of light alkane and other compounds by iron photocatalysis. *Nat. Catal.* 4, 1487–1492 (2021).

21. Li, Z., Wang, X., Xia, S. & Jin, J. Ligand-activated iron photocatalysis enabling deoxyalkylation of alkylcations. *Org. Lett.* 21, 4259–4265 (2019).

22. Xie, S., Hu, K., Lei, C. & Jin, J. Intramolecular aromatic C-H acylation enabled by iron photocatalysis. *Org. Lett.* 22, 1385–1389 (2020).

23. Garza-Sanchez, R. A., Tlahuext-Aca, A., Tavakoli, G. & Glorius, F. Visible light-mediated direct decarbonylation C–C functionalization of heteroarenes. *ACS Catal.* 7, 4057–4061 (2017).

24. Crespi, S. & Fagnoni, M. Generation of alkyl radicals from the tyranny of tin to the photon democracy. *Chem. Rev.* 120, 9790–9833 (2020).

25. Li, N. L. et al. A highly selective decarbonylation deuteriation of carboxylic acids. *Chem. Sci.* 12, 5505–5510 (2021).

26. Zhang, L., Si, X., Rominger, F. & Hashmi, A. S. K. Visible-light-induced radical carbo-cyclization/gem-diboration through triplet energy transfer between a gold catalyst and aryliodides. *J. Am. Chem. Soc.* 142, 10485–10493 (2020).

27. Zhao, H., Schmidt, L. C., Bonin, J. & Robert, M. Visible-light-driven methane formation from CO2 with a molecular iron catalyst. *Nature* 548, 74–77 (2017).

28. Hu, Y. et al. Next-generation D2-symmetric chiral porphyrins for Cobalt(II)-based metaloradical catalysis: catalyst engineering by distal bridging. *Angew. Chem. Int. Ed.* 58, 2670–2674 (2019).

29. Hu, Y. et al. Enantioselective radical construction of 5-membered cyclic sulfonamides by metaloradical C-H activation. *J. Am. Chem. Soc.* 141, 20802–20806 (2019).
Acknowledgements

We thank the National Natural Science Foundation of China (22122103 to J.X., 22101130 to J.H., 21971111 to C.Z., 21971108 to J.X., 21732003 to C.Z.), the Natural Science Foundation of Jiangsu Province (Grant No. BK20190006 to J.X.), the Fundamental Research Funds for the Central Universities (20Z514380214 to J.X.), China Postdoctoral Science Foundation (2020TQ0139 and 2021M701661 to C.-G.Z.), Foundation of Advanced Catalytic Engineering Research Center of the Ministry of Education of Hunan University (No. 2020AC001 to J.X.), and “Innovation & Entrepreneurship Talents Plan” of Jiangsu Province. Zhi Liu, Tao Zhong, and Bo Ling are warmly acknowledged for their help to reproduce experimental procedures for products (23, 14, and 55).

Author contributions

J.X. and S.W. conceived and designed the project. S.W. and T.L. performed and analyzed the experimental data. J.X. wrote the manuscript with input from all authors and discussed the manuscript with C.G., J.H., C.-G.Z., C.Z., and H.T.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41467-022-30176-z.

Correspondence and requests for materials should be addressed to Chengjian Zhu or Jin Xie.

Peer review information Nature Communications thanks the anonymous reviewers for their contribution to the peer review of this work.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2022