Adsorption-supported ozonation of complex mariculture wastewater pharmaceuticals by greener (Ni/Fe-C) nano-composite derived by co-pyrolysis of Coconut Shell & partially reduced iron: mechanism, and implications

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Text S1. Chemicals, materials, and nano-composites synthesis

Oxytetracycline ($C_{22}H_{24}N_2O_9$) (79-57-2) ⩾99%, HPLC was purchased from Aladdin Macklin Biochemical Co., Ltd. (Shanghai, China). Coconut shell residues were obtained after food waste at a plant at the Xiamen Eastern Solid Waste Treatment Center. Partially reducing iron ore waste was collected from pig iron manufacturing units. Dimethyl sulfoxide (C2H6OS F.W. 78.13, ⩾99.5%), tert-butanol (C4H10O F.W. 74.12⩾99.5%), 2,2,6,6-tetramethylpiperidine (TEMP, ⩾ 98.0%), and 5 5-dimethyl-1-pyrroline-N-oxide (DMPO, ⩾97.0%) were purchased from Shanghai Maclean's Biochemical Technology Co., Ltd. In addition, 1-4 Benzoquinone (BQ ⩾ 97.0%) was purchased from Tokyo Chemical Industry Co., Ltd. Sodium chloride NaCl (F.W. 58.44, ⩾99.5%). All chemical reagents employed in this study were analytically pure. Super deionized water (SDI) was used in a Milli-Q Biocel water system, and the oven was purchased from Shanghai Jinghong Experimental Equipment Co., Ltd. All pieces of glassware were washed with SDI water several times before being used and then dried in an oven at 105 °C.

Nano-composite Synthesis: Co-pyrolysis was carried out in a horizontal tube furnace (OTF-1200X, MTI Corp.) with a 60 mm O.D. quartz tube. A 5 g portion of the Coconut shell residue (CNR) and 1 g reduced iron (RI) powder (5:1 w/w) was spread as a thin layer (ca. 1 mm) on a quartz boat and placed in the constant temperature zone of the tube. The samples were heated from room temperature to 300-700 °C at 5 °C min^{-1} under flowing N_2 (200 mL min^{-1}, 99.99 % purity). The temperature was held for 45 min, then the furnace was shut off and allowed to cool naturally to below 60 °C under flowing N_2 (200 mL min^{-1}). Targets of 300 °C and 500 °C were run with the same heating rate, hold time, and gas flow.

Reproducibility:

The entire 300-700 °C run was repeated three times (on different days, using different 5 g of CNR and 1 g of RI scoops from the well-mixed stock) to demonstrate reproducibility, with the following results (mean ± uncertainty): solid yield 30.2 ± 1.4 % and Fe content 10.1 ± 0.2 wt % (ICP-OES). Bed temperature was within ±3 °C of 300-700 °C for all three runs, N_2 flow drift was <1 %, and outlet O_2 was < 10 ppm.

Run ID	Solid yield (%)	Fe (wt%)
R-700-1	25.3	16.4
R-700-2	24.8	16.2
R-700-3	25.2	16.3
Mean ± SD	25.1 ± 0.3	16.3 ± 0.1

*yield = (final mass / 6 g) × 100 %.

Text S2: Catalytic Characterization and Analytical Method

The physiological morphology and elemental composition of all SCS@BR catalysts were examined using advanced technologies, including Energy Dispersive Spectroscopy (E.D.S.), Scanning electron microscopy (S.E.M., S-4800, Hitachi, Japan), and a transmission electron microscope (T.E.M., JEM1200EX, J.E.O.L., Japan). ASAP 2020. X-ray photoelectron spectroscopy (XPS) was used to analyze the chemical environment before and after the reaction. The instrument used for this analysis was the Axis Supra XPS and X-ray Fluorescence Spectrometer system manufactured by Shimadzu in Japan. To know about the TOC values, catalytically degraded samples were analyzed by a TOC analyzer (TOC－LCPH, Shimadzu,

Japan). COD values of all samples were determined by utilizing COD (HJ/T 399-2007) with an automatic digester by Shandong Horde Electronic Technology Co., Ltd. A ball mill (XQM-4; Changsha Tian-chuang Power Technology, China) was used.

The total organic elimination efficiency following the reaction was measured using a T.O.C. (Japanese Shimadzu) instrument. In addition, identifying potential reactive oxygen species (R.O.S.) created for mechanism research was conducted utilizing a Bruker Electron Spin Resonance spectrometer (ESR). A powder diffraction meter (XRD, X 'Pert Pro, P.A.N. analytical, Netherlands) was equipped with Cu-Kα radiation (λ = 1.5444426 Å) to determine the crystallinity of the Fe-BRB modified. Fourier transform infrared spectroscopy (FT-IR) was conducted using the KBr pellet technique over 4000 to 400 cm^{-1}. LC-MS was used to determine the OFL intermediate products throughout the catalytic degradation reaction, thereby uncovering the potential pathway of OFL degradation.

Analytical methods:

Catalytic activity calculations: The following equations calculate the degradation efficiency/catalytic activity of OTC

$$Degradation\ efficiency\ of\ OTC\ (\%) = [\frac{Ct - Co}{Co}] \times 100\% \quad (1)$$

C_t is the concentration at a particular time (min), and C_o is the initial concentration of OTC. COD is the value of the chemical oxygen demand at time (t=0) and time (t) of all samples.

$$Degradation\ efficiency\ of\ COD\ (\%) = [\frac{CODt - CODo}{CODo}] \times 100\% \quad (2)$$

All optimized catalytic ozonation degradation experiments were performed and repeated, and then the average values were taken. Additionally, standard deviation (SD) values

for OTC over SDI water and stimulated urine experimental values were taken for repeated experimental values.

$$SD_{mean} = \sqrt{\frac{{SD_1}^2_{try} + {SD_2}^2_{try}}{2}} \quad (3)$$

${SD_1}^2_{try}$ and ${SD_2}^2_{try}$ It is the standard deviation of repeated experiments.

Further, standard error (SE) values were also calculated using the following values. “n” represents the number of samples taken over 100 min of catalytic reaction.

$$SE = \frac{SD}{\sqrt{n}} \quad (4)$$

All DFT work was carried out in **Materials Studio 2020** with the **DMol³** module. We chose the **PBE GGA** functional plus the **TS dispersion correction** to capture long-range forces, used the numerical double-ζ **polarized DNP 4.4** basis set within a **3.5 Å** cutoff, and kept **core electrons frozen** with the **DFT semi-core pseudopotential.** The model was a single-layer graphene flake whose zig-zag edge carried one substitutional Ni or Fe atom; a **15 Å** vacuum slab prevented periodic images. Optimizations converged to forces below **0.004 Ha Å⁻¹** and energy changes below **1 × 10⁻⁵ Ha**. Ozone was docked side-on to the metal–carbon bond, and reaction paths were traced with the **LST/QST algorithm,** each transition-state verified by one imaginary frequency; spin polarization was retained throughout.

Text. S3: Escherichia coli (E. coli) growth, Chinese cabbage growth, and Zebra-fish embryo assessment over SDI, polluted and treated wastewater samples

Chinese cabbage grain growth: For the germination of the Chinese cabbage, healthy grains were put into three different plates having SDI, contaminated water, and treated water after cleaning and washing, for 80 days, and the temperature was maintained at 30°C.

Escherichia coli (E. coli) growth:

Materials Required:

All solutions were prepared using SDI water. The Luria Bertani (LB) broth was made by dissolving 10 g of LB powder in 500 mL of water. A saline solution was prepared by dissolving 20 g of NaCl per liter of water. Agar-agar powder (8 g) was added to 500 mL of LB broth to create solid LB agar media. For the buffer solution, sodium dihydrogen phosphate (NaH_2PO_4) and disodium phosphate (Na_2HPO_4) were used in a 1:1 ratio. The equipment used included an incubator, a Bunsen burner or alcohol lamp (for sterilizing inoculating loops), and an E. coli bacterial culture.

Preparation of Solid Media Plates:

Escherichia coli was cultured by inoculating a known working stock into LB liquid medium and incubating at 30 °C for 12 h. The culture was centrifuged at 8,000 rpm for 10 min, the supernatant removed, and the pellet resuspended in sterile buffer to an OD_{600} of 2. Agar plates were made by dissolving the medium, autoclaving at 121 °C for 30 min, cooling to 50 °C, and pouring aseptically into sterile Petri dishes. For enumeration, serial dilutions were made in PBS (eight-fold, 10^{-1} to 10^{-8}) by transferring 1 µL of culture into 9 µL of sterile PBS, mixing well, and plating 0.1 mL of each dilution onto the agar. After incubation at 37 °C for 24 h, colonies were counted before and after antibiotic exposure to determine treatment efficacy. In parallel phytotoxicity tests, agricultural soil was sown with 5 g of wheat grains and irrigated for 5 days with (i) sterile distilled water (SDI), (ii) raw mariculture wastewater, (iii) untreated wastewater, and (iv) ozonation-treated wastewater; significant differences in germination and shoot length were noted.

Zebra fish embryo growth:

Zebrafish embryos, collected within 2 hours post-fertilization (hpf), were maintained in E3

medium at 28 °C. At 24 hpf, enzymatic dechorionation was performed using pronase, followed by anaesthesia with 0.02 mg ml^{-1} tricaine at 48 hpf. Subsequently, the embryos were divided into three distinct treatment groups: (i) ozonation-treated mariculture water, (ii) untreated mariculture water, and (iii) sterile distilled water (SDI). Incubation proceeded at 28 °C, with observations conducted every 24 hours. At 120 hpf, key parameters including hatching success, heart rate, axial curvature, and total length were documented and statistically compared across the treatments, ensuring compliance with an approved ethical protocol.

Test S4. Avoided Burdens from Waste Utilization (Waste-Based Catalyst Preparation)

Avoided-burden equation (waste-based catalyst)

Avoided $Impact_i$ [kg CO_2-eq or PO_4-eq] = m_waste [kg] × CF_disposal,i [kg_waste^{-1} kg CO_2-eq] [1] , [2]

Description

- *m*waste: mass of coconut-shell residue or partially-reduced iron ore waste diverted from disposal and used in the composite (kg).
- CFdisposal,*i*: characterization factor for the avoided disposal route (landfill, incineration, etc.) and impact category *i* (GWP, eutrophication, etc.).
- Product of the two terms quantifies the environmental credit gained by not landfilling or incinerating the waste.

2. Toxicity from Potential Impurities in Waste Feedstock (Catalyst Preparation & Usage)

Toxicity-impact equation (impurities from waste feedstock)

$$Impact_{toxicity,\,j}\ [CTUh] = k \sum m\text{toxic},k\ [kg] \times CF_{toxicity,j,k}\ [kgCTUh]$$

[3] [4]

What each term means

- *Impact* – total toxicity score for category *j* (human toxicity, freshwater ecotoxicity, etc.).
- *m* – mass of impurity *k* (heavy metal, POP, etc.) released during catalyst preparation or use.
- *CF* – characterization factor that converts kilograms of substance *k* into comparative toxic units (CTU), accounting for fate, exposure, and effect.

3. Resource Efficiency & Circularity Metrics (Catalyst Life Cycle)

Material Circularity Indicator (MCI) – Ellen MacArthur Foundation [5], [6], [7].

MCI=(1−V) × ((1-Fm)/ (Fw+Fc))

Table

Symbol	**Meaning (fraction or ratio)**
V	Virgin material input per functional unit (kg virgin/kg product)
F<sub>	Share of **recycled material** in the **feedstock** (mass%)
F<sub>	Share of **virgin material** in the **feedstock** (mass% %)
F<sub>	Share of **recycled content** in the **final product** (mass% %)

Interpretation:

MCI = 1 → **perfect circularity** (no virgin input, fully recycled).

MCI = 0 → **linear system** (all virgin, no recycling).

4. Regeneration Efficiency and Impact (Catalyst Discharge/End-of-Life)

Regeneration credit per functional cycle

Credit reg, *i* [Cycle kg CO2-eq] = (Impact fresh, *I* – Impact reg, *i*)×Number of Cycles

[8]

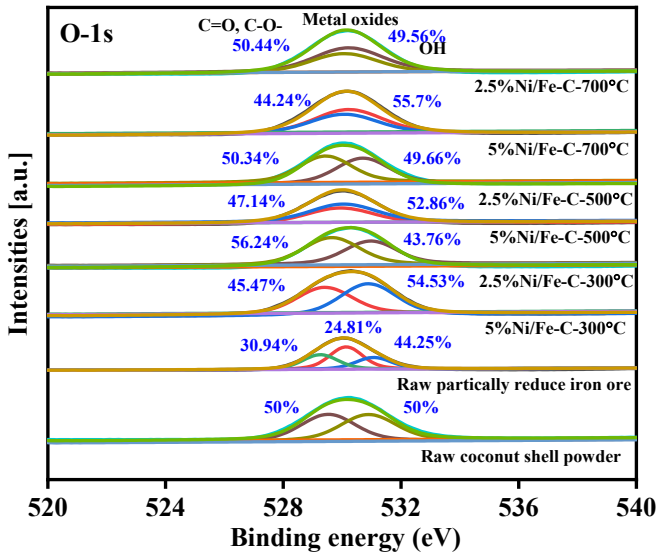
Term	**Meaning**
Impact<sub>	Life-cycle impact of producing a **virgin** catalyst for category *i* (e.g., GWP, CO_2-eq).
Impact<sub>	Life-cycle impact of **one regeneration** cycle (energy, chemicals, emissions) for the same category.
Number of Cycles	Completed regeneration loops before catalyst discard.

Interpretation:

The equation quantifies the **net environmental saving** obtained by regenerating the waste-derived catalyst instead of repeatedly manufacturing fresh material.

Figure S1: O-1s peaks of all bimetallic (Fe/Ni) nano-composites

(a)



(b)

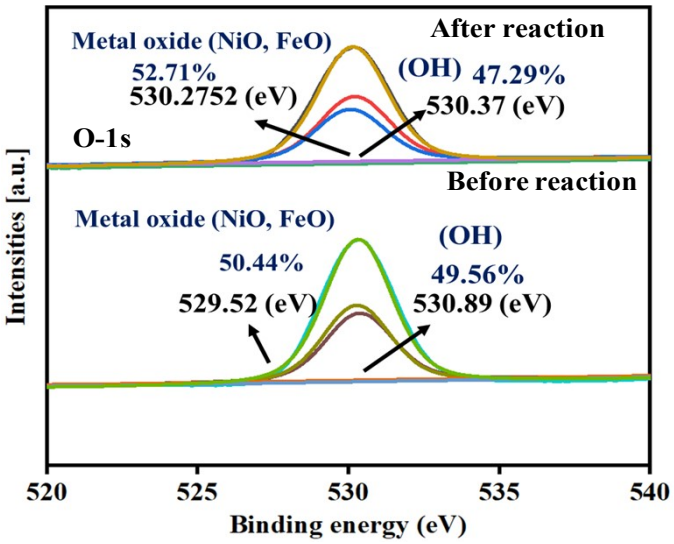


Figure S2: 3D-EEMs OTC treatment before and after reaction

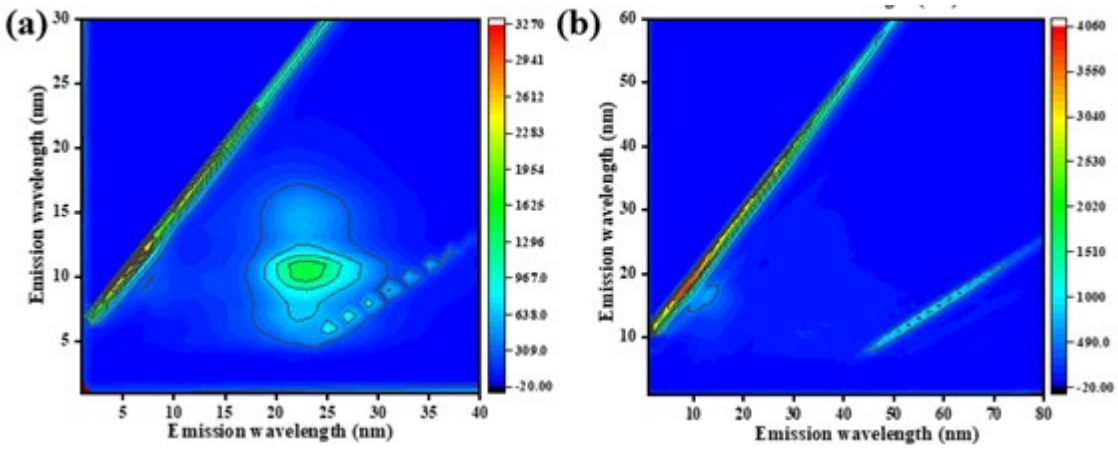


Figure S3: DMPO singles trapped superoxide radicals ($O_2^{\bullet-}$)

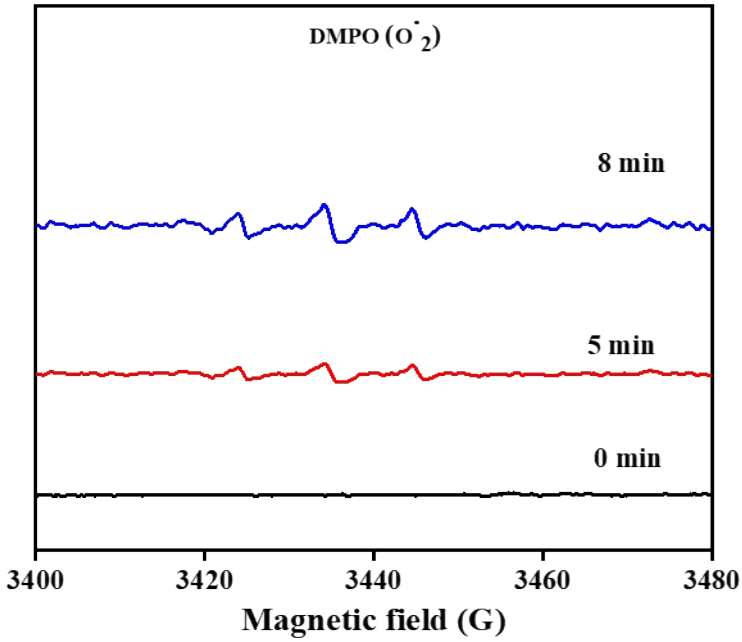


Figure S4: Performance comparison

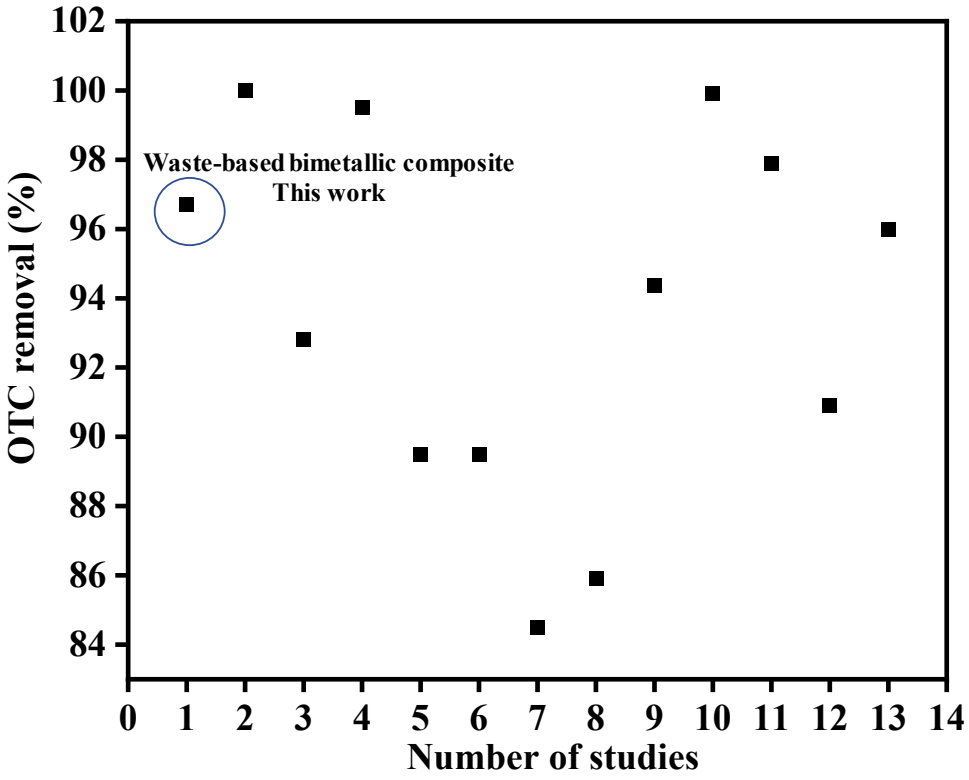
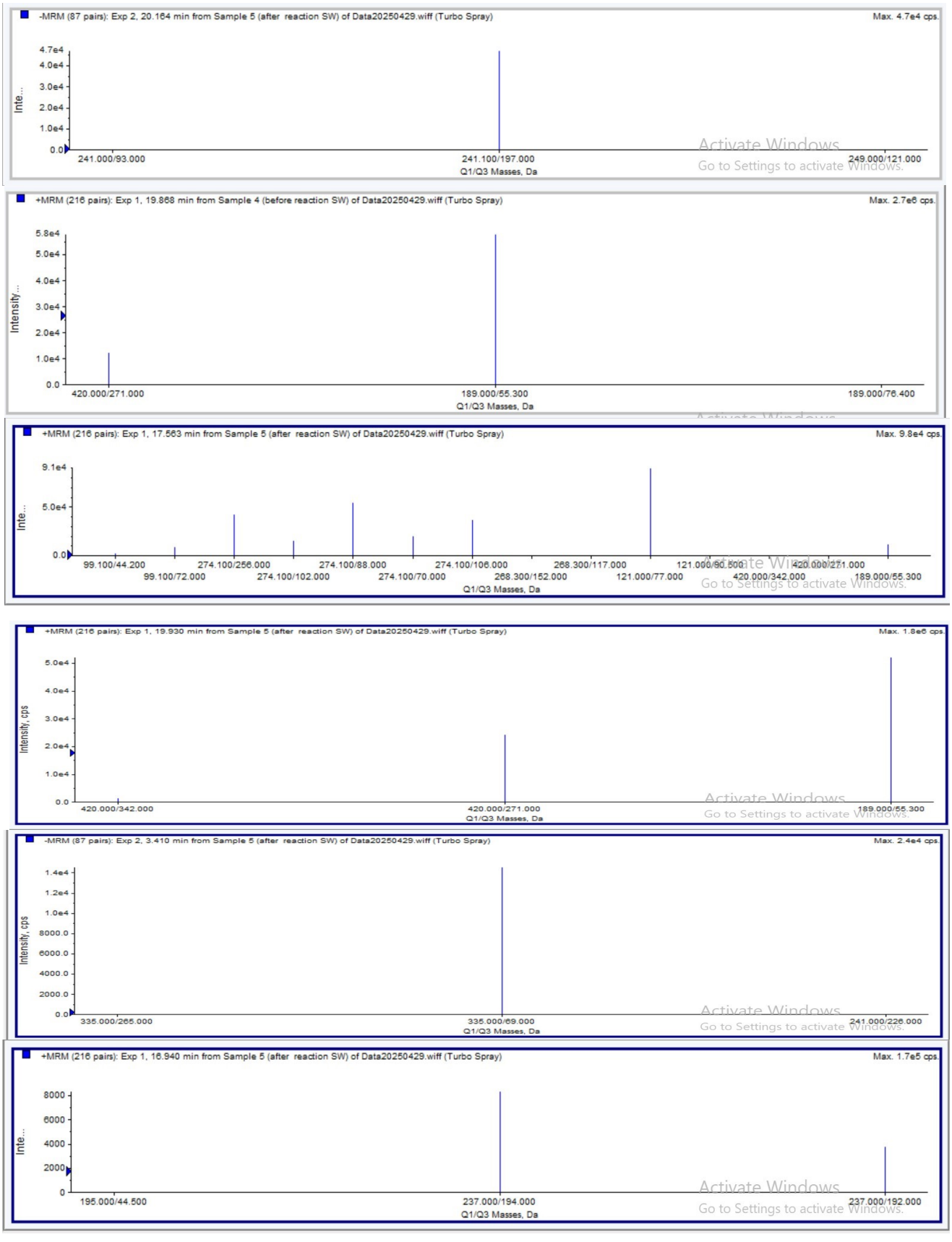
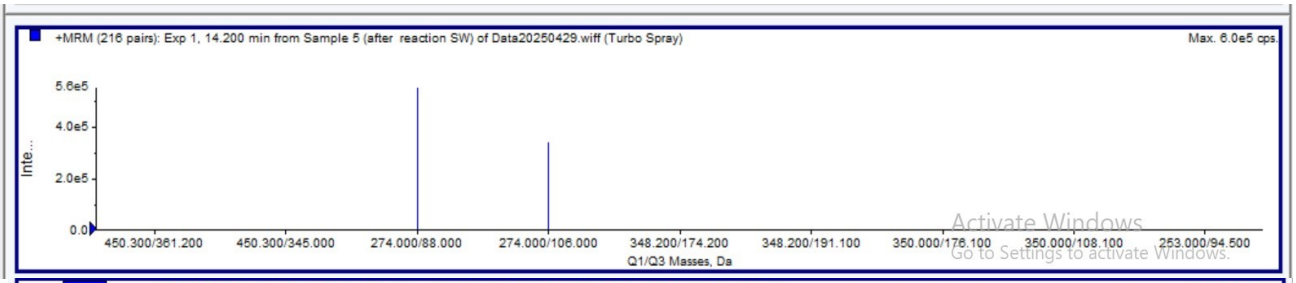
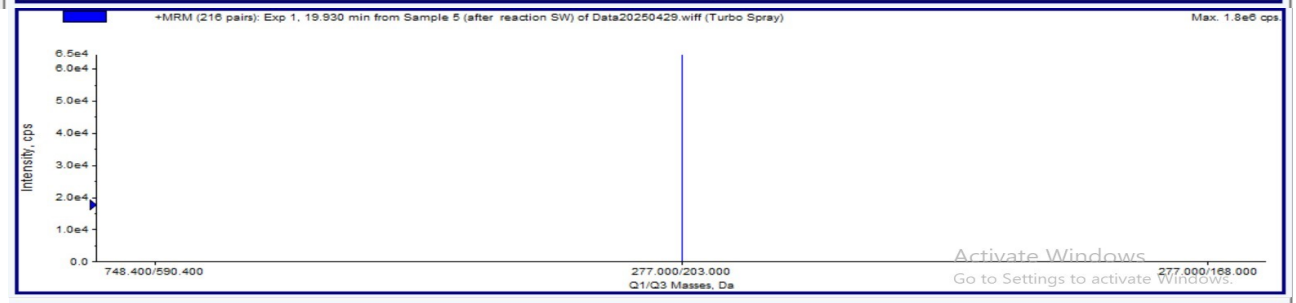
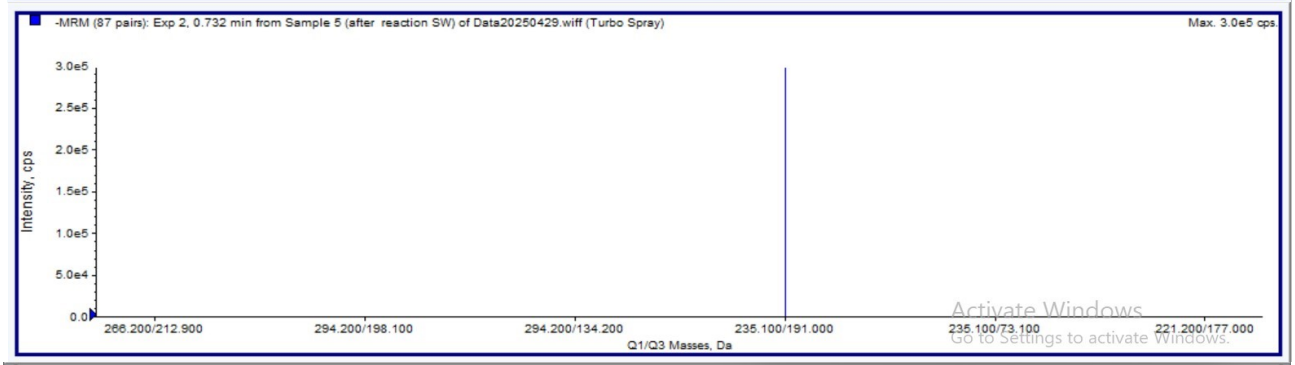


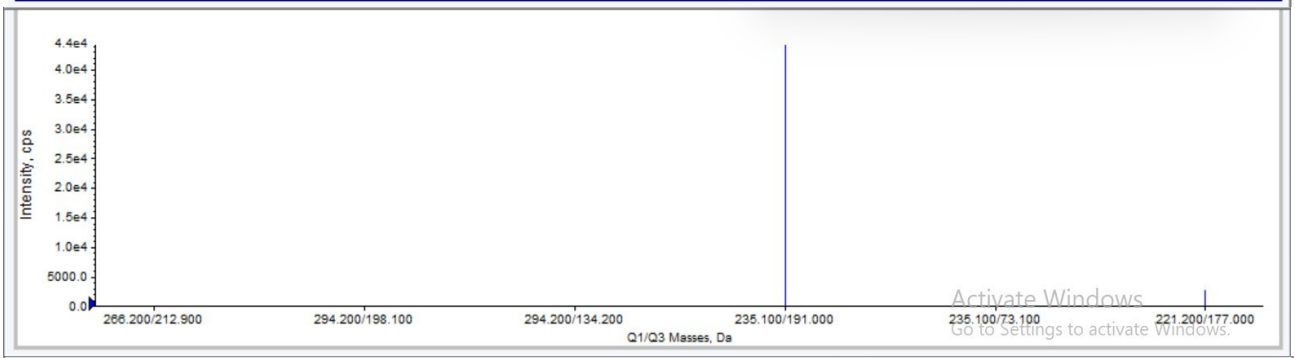
Figure S5: Mariculture wastewater toxic pharmaceuticals (Before reaction)

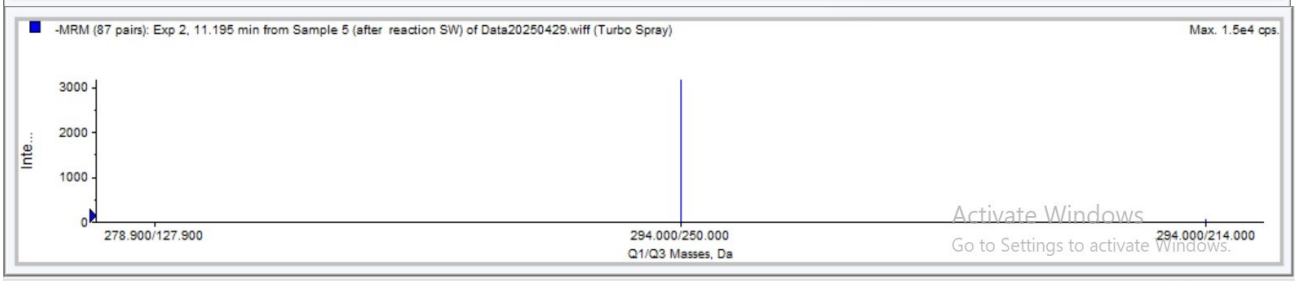


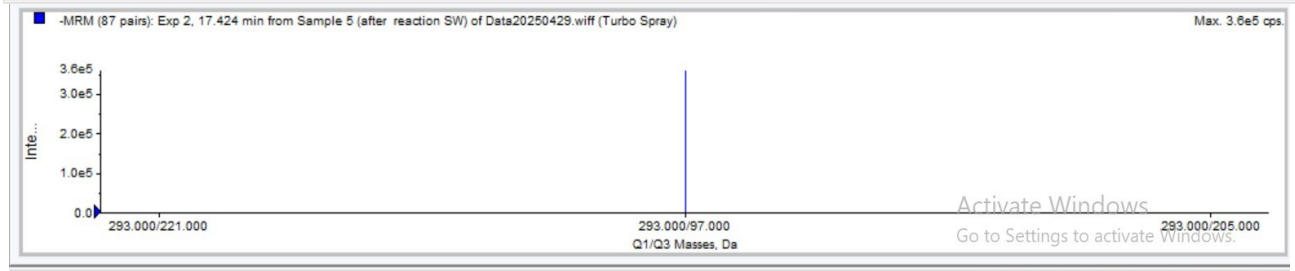

+MRM (216 pairs): Exp 1, 14.200 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 6.0e5 cps.
Inte...
5.6e5
4.0e5
2.0e5
0.0
450.300/361.200
450.300/345.000
274.000/88.000
274.000/106.000
348.200/174.200
348.200/191.100
350.000/176.100
350.000/108.100
253.000/94.500
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

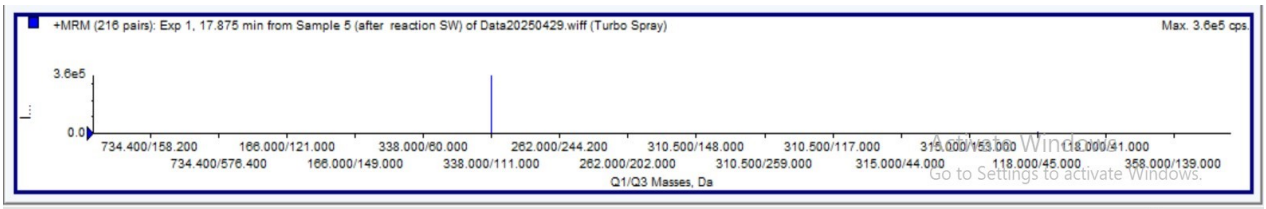

+MRM (216 pairs): Exp 1, 19.930 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.8e6 cps.
Intensity, cps
6.5e4
6.0e4
5.0e4
4.0e4
3.0e4
2.0e4
1.0e4
0.0
748.400/590.400
277.000/203.000
277.000/168.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

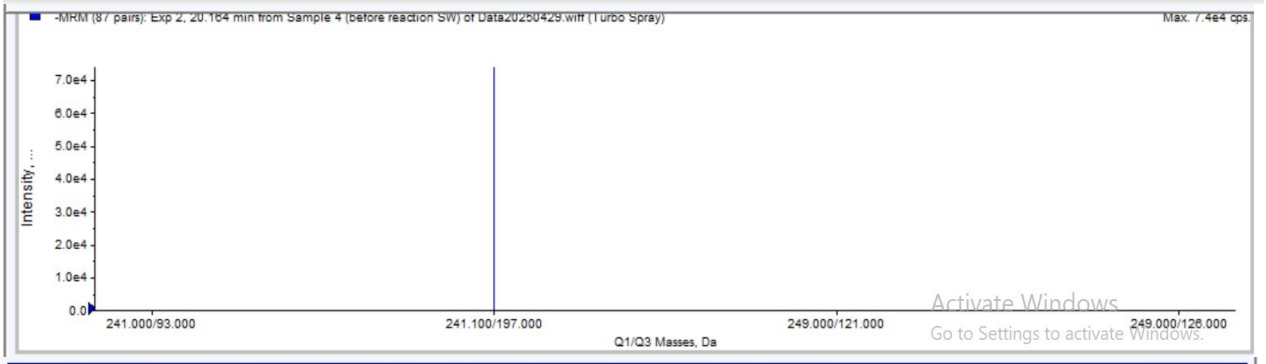

-MRM (87 pairs): Exp 2, 0.732 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 3.0e5 cps.
Intensity, cps
3.0e5
2.5e5
2.0e5
1.5e5
1.0e5
5.0e4
0.0
266.200/212.900
294.200/198.100
294.200/134.200
235.100/191.000
235.100/73.100
221.200/177.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

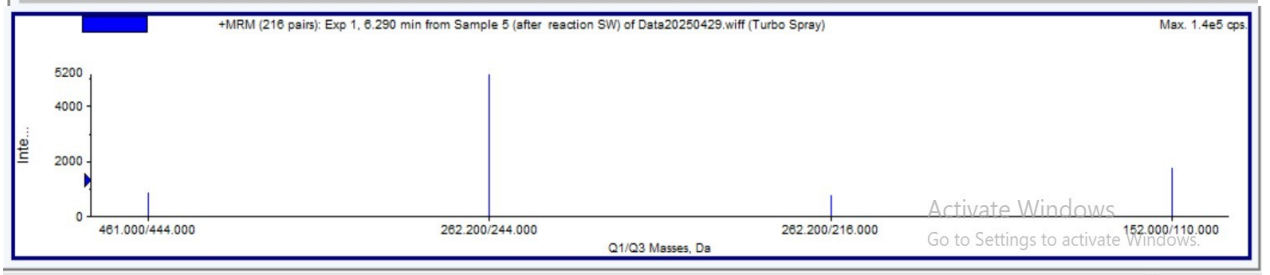

Intensity, cps
4.4e4
4.0e4
3.5e4
3.0e4
2.5e4
2.0e4
1.5e4
1.0e4
5000.0
0.0
266.200/212.900
294.200/198.100
294.200/134.200
235.100/191.000
235.100/73.100
221.200/177.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

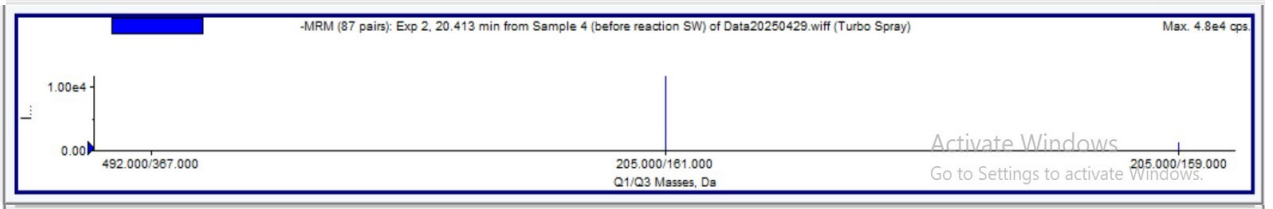

-MRM (87 pairs): Exp 2, 11.195 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.5e4 cps.
Inte...
3000
2000
1000
0
278.900/127.900
294.000/250.000
294.000/214.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

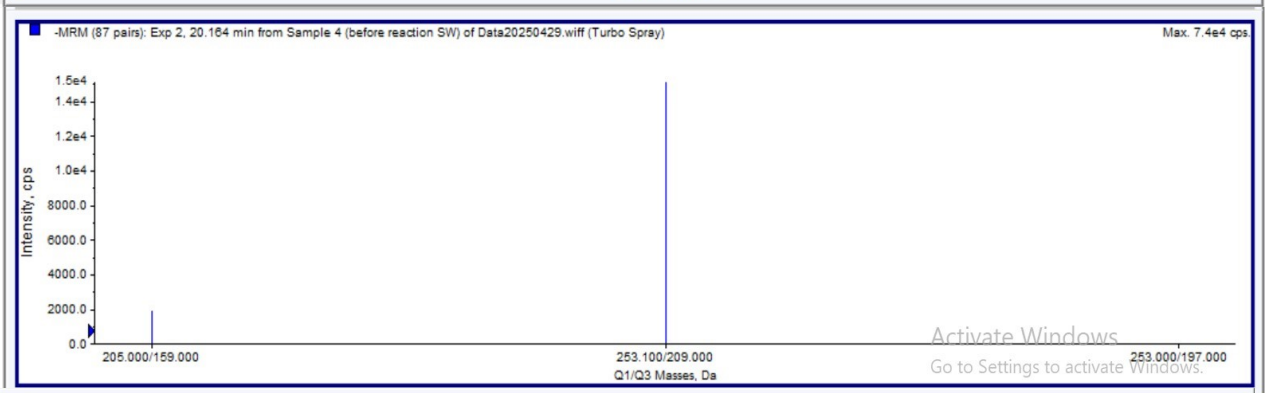

-MRM (87 pairs): Exp 2, 17.424 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 3.6e5 cps.
Inte...
3.6e5
3.0e5
2.0e5
1.0e5
0.0
293.000/221.000
293.000/97.000
293.000/205.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

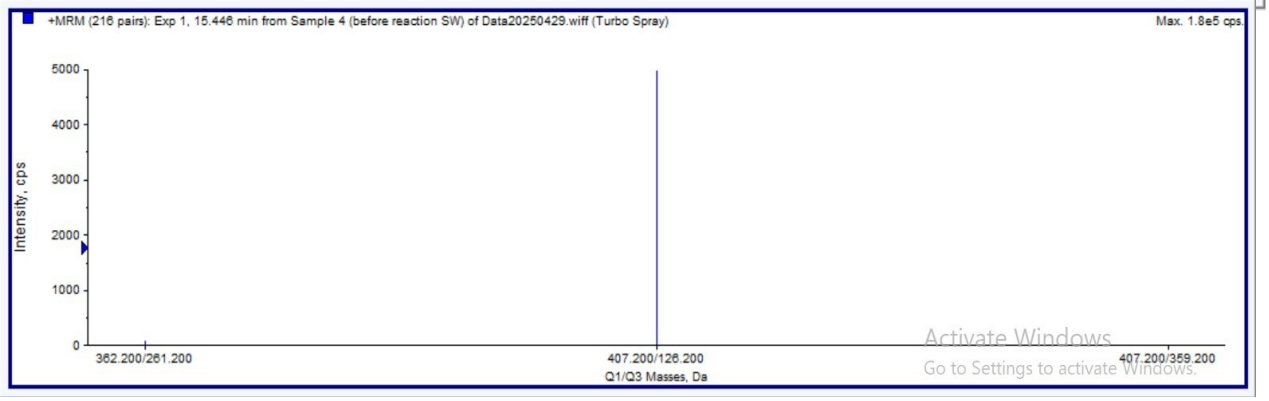

+MRM (216 pairs): Exp 1, 17.875 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 3.6e5 cps.
3.6e5
0.0
734.400/158.200
166.000/121.000
338.000/60.000
262.000/244.200
310.500/148.000
310.500/117.000
315.000/153.000
118.000/41.000
734.400/576.400
166.000/149.000
338.000/111.000
262.000/202.000
310.500/259.000
315.000/44.000
118.000/45.000
358.000/139.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


-MRM (87 pairs): Exp 2, 20.164 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 7.4e4 cps.
Intensity, ...
7.0e4
6.0e4
5.0e4
4.0e4
3.0e4
2.0e4
1.0e4
0.0
241.000/93.000
241.100/197.000
249.000/121.000
249.000/126.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 6.290 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.4e5 cps.
Inte...
5200
4000
2000
0
461.000/444.000
262.200/244.000
262.200/216.000
152.000/110.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


-MRM (87 pairs): Exp 2, 20.413 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 4.8e4 cps.
I...
1.00e4
0.00
492.000/367.000
205.000/161.000
205.000/159.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


-MRM (87 pairs): Exp 2, 20.164 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 7.4e4 cps.
Intensity, cps
1.5e4
1.4e4
1.2e4
1.0e4
8000.0
6000.0
4000.0
2000.0
0.0
205.000/159.000
253.100/209.000
253.000/197.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.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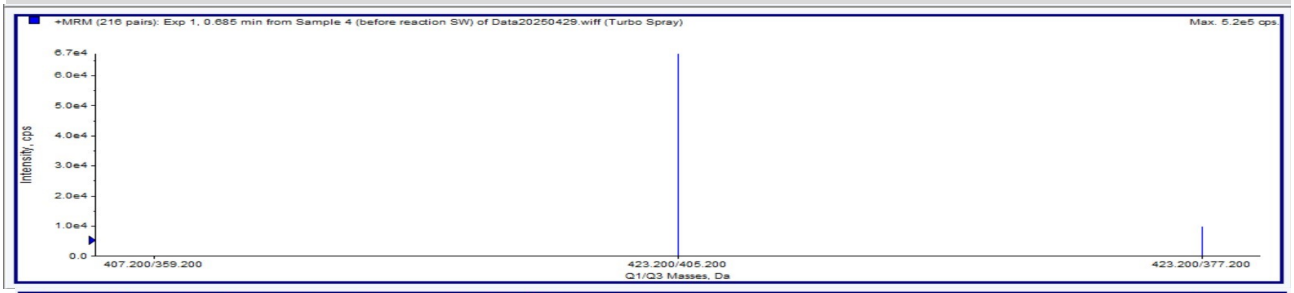
Max. 1.8e5 cps.
Intensity, cps
5000
4000
3000
2000
1000
0
362.200/261.200
407.200/126.200
407.200/359.200
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.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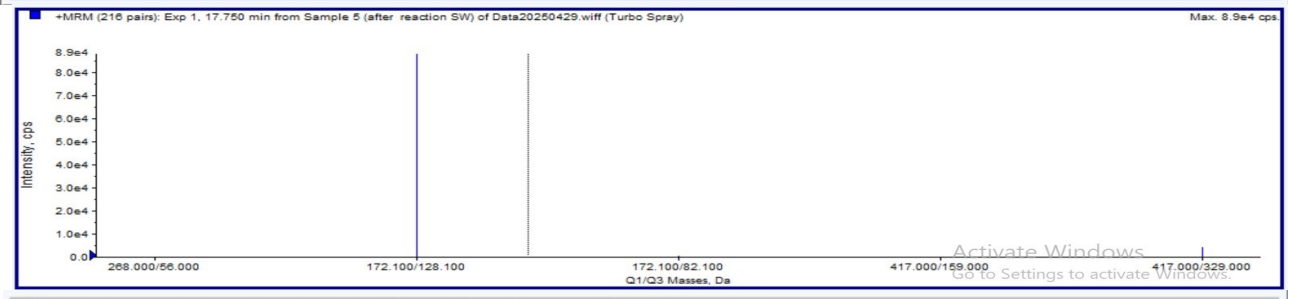
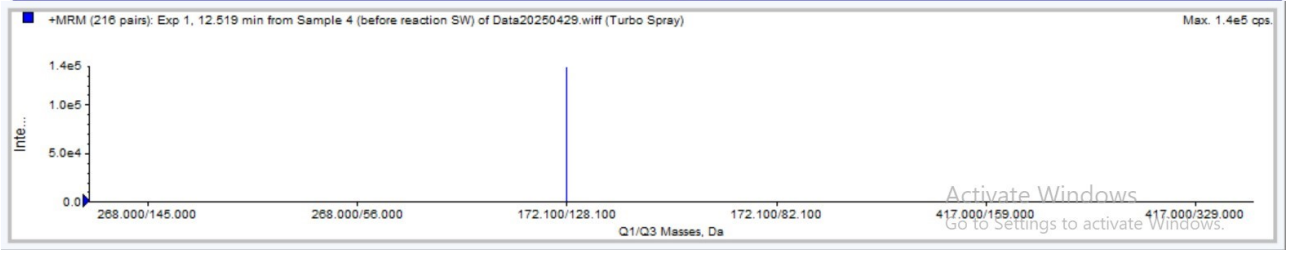
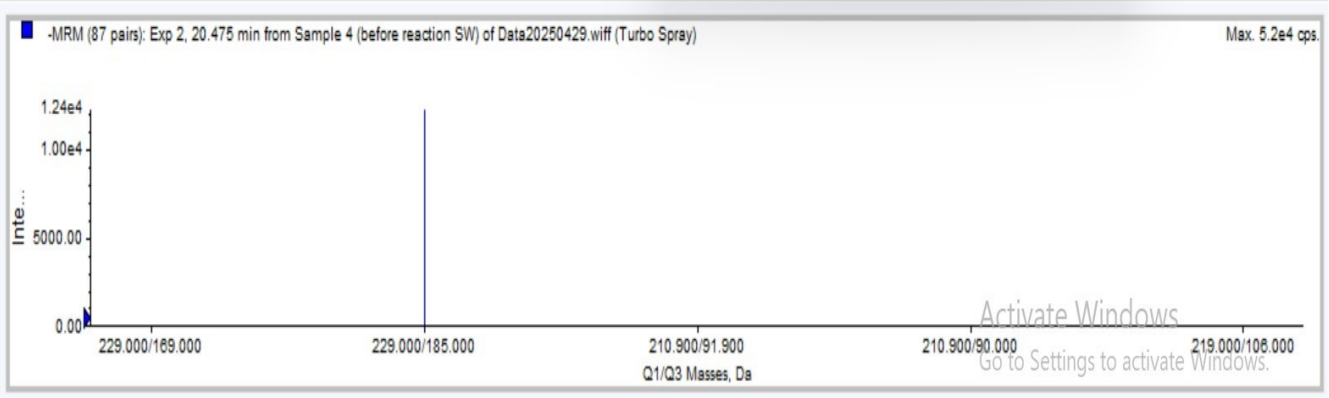
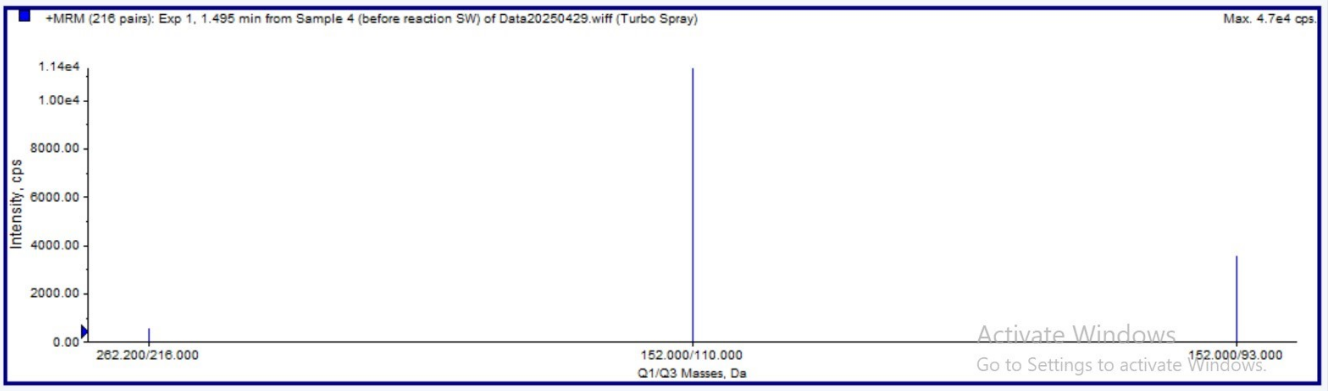
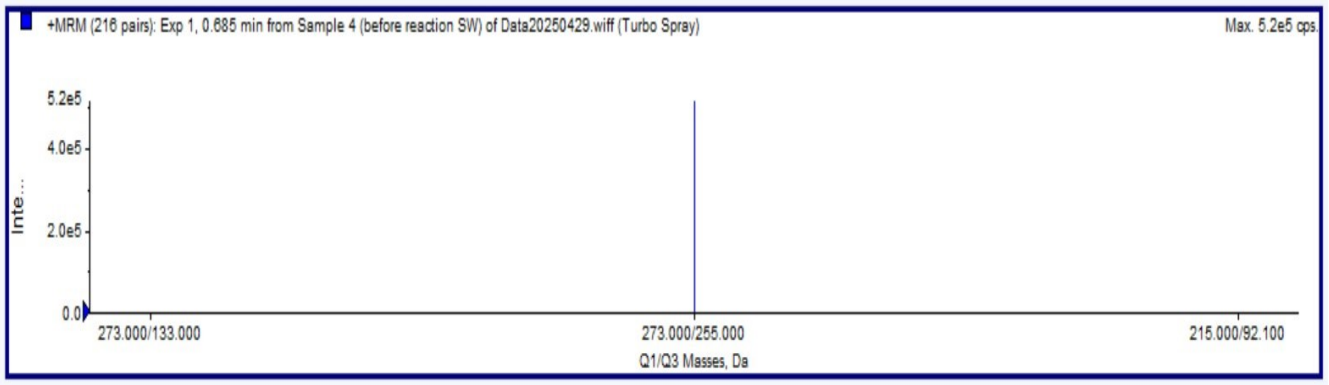
Max. 5.2e5 cps.
Intensity, cps
6.7e4
6.0e4
5.0e4
4.0e4
3.0e4
2.0e4
1.0e4
0.0
407.200/359.200
423.200/405.200
423.200/377.200
Q1/Q3 Masses, Da


+MRM (216 pairs): Exp 1, 17.750 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 8.9e4 cps.
Intensity, cps
8.9e4
8.0e4
7.0e4
6.0e4
5.0e4
4.0e4
3.0e4
2.0e4
1.0e4
0.0
268.000/56.000
172.100/128.100
172.100/82.100
417.000/159.000
417.000/329.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 12.519 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.4e5 cps.
Inte...
1.4e5
1.0e5
5.0e4
0.0
268.000/145.000
268.000/56.000
172.100/128.100
172.100/82.100
417.000/159.000
417.000/329.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


-MRM (87 pairs): Exp 2, 20.475 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 5.2e4 cps.
1.24e4
1.00e4
5000.00
0.00
Inte...
229.000/169.000
229.000/185.000
210.900/91.900
210.900/90.000
219.000/106.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 1.495 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 4.7e4 cps.
1.14e4
1.00e4
8000.00
6000.00
4000.00
2000.00
0.00
Intensity, cps
262.200/216.000
152.000/110.000
152.000/93.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 0.685 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 5.2e5 cps.
5.2e5
4.0e5
2.0e5
0.0
Inte...
273.000/133.000
273.000/255.000
215.000/92.100
Q1/Q3 Masses, Da

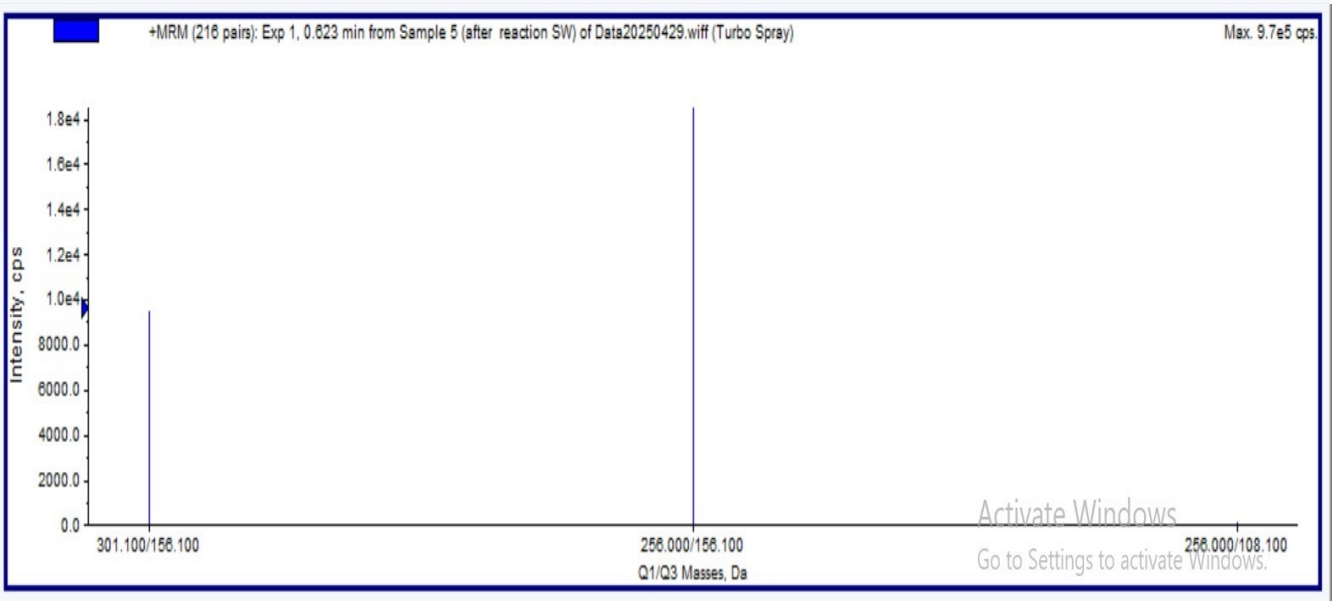
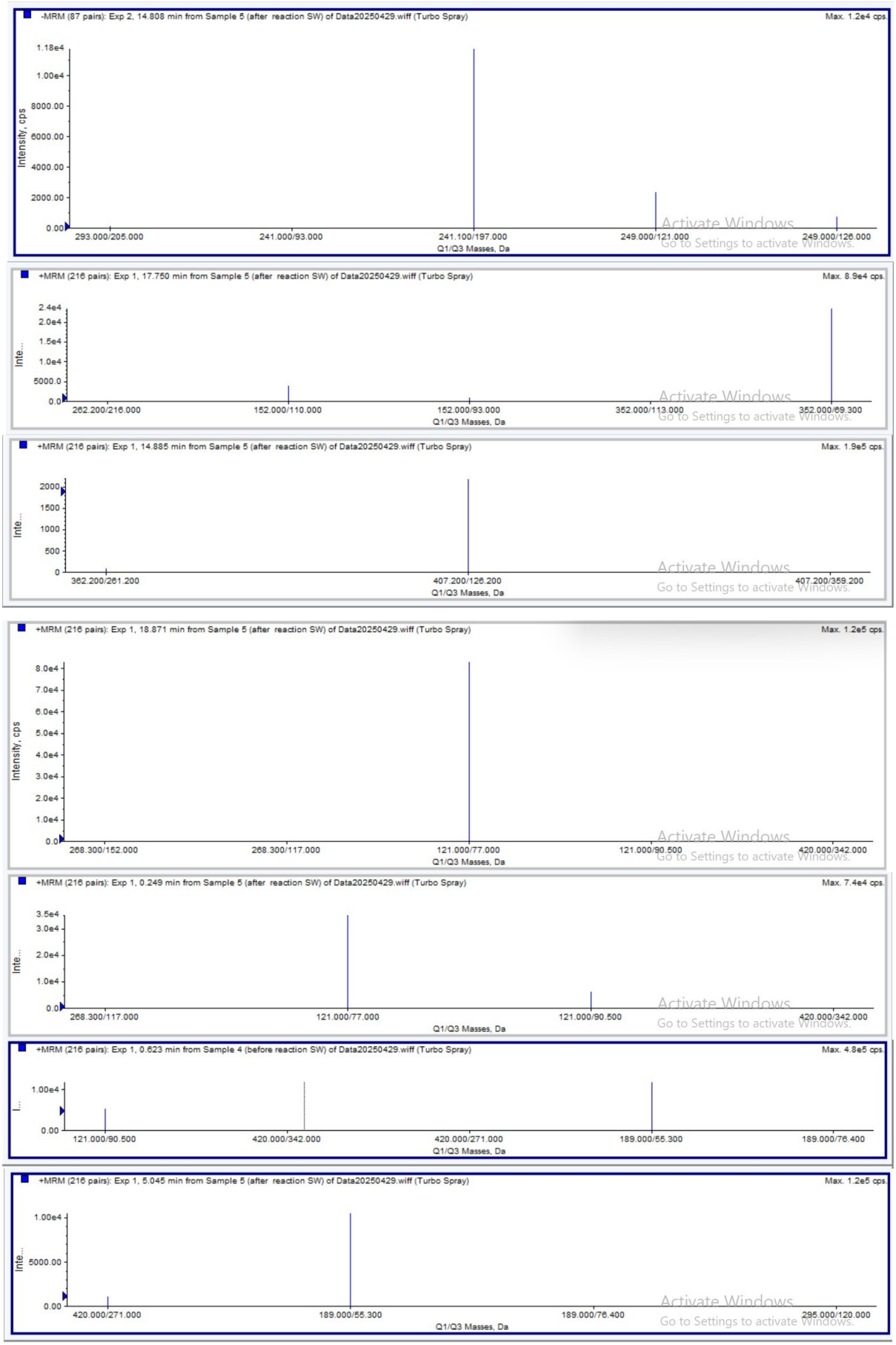
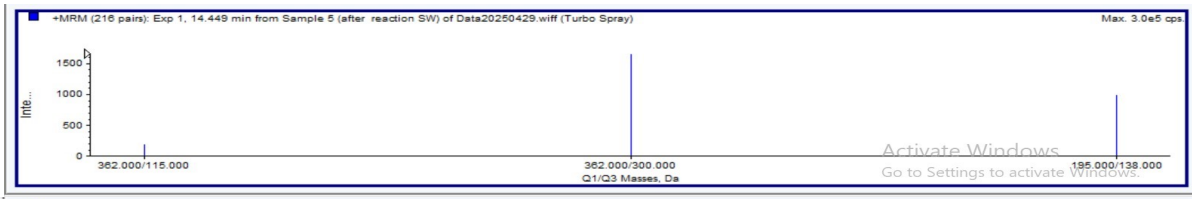
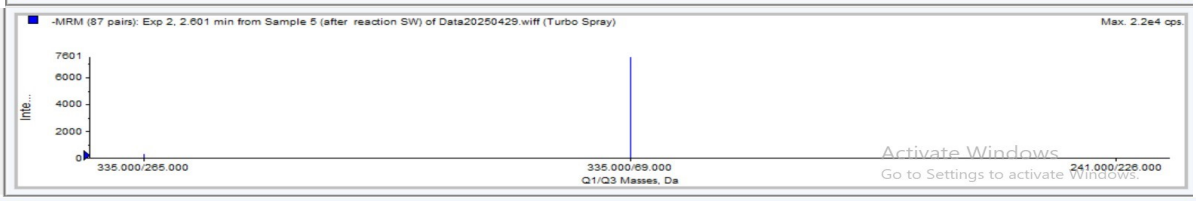
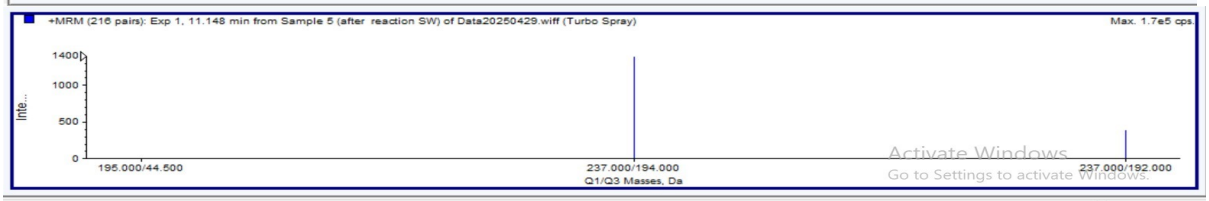

+MRM (216 pairs): Exp 1, 0.623 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 9.7e5 cps.
1.8e4
1.6e4
1.4e4
1.2e4
1.0e4
8000.0
6000.0
4000.0
2000.0
0.0
Intensity, cps
301.100/156.100
256.000/156.100
256.000/108.100
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

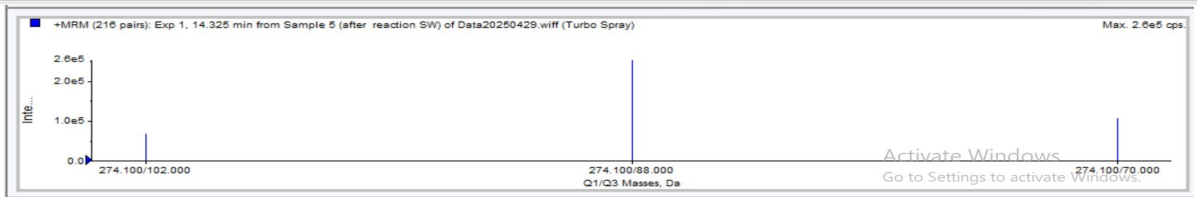
Figure S6: Mariculture wastewater toxic pharmaceuticals (After reaction)

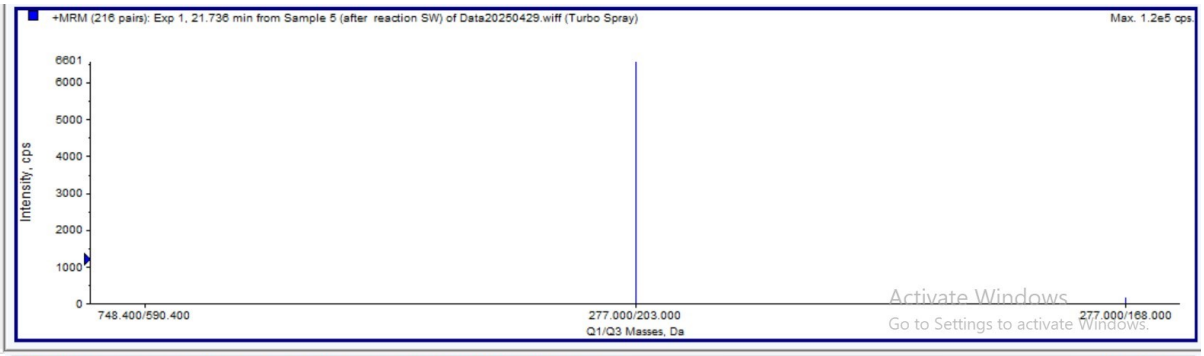


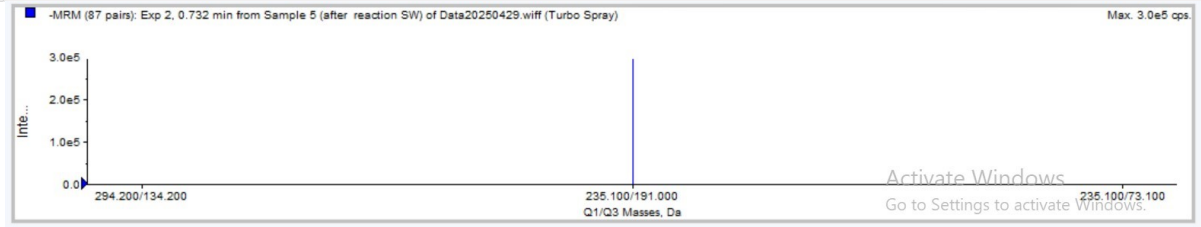

+MRM (216 pairs): Exp 1, 14.449 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 3.0e5 cps.
Inte...
1500
1000
500
0
362.000/115.000
362.000/300.000
195.000/138.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

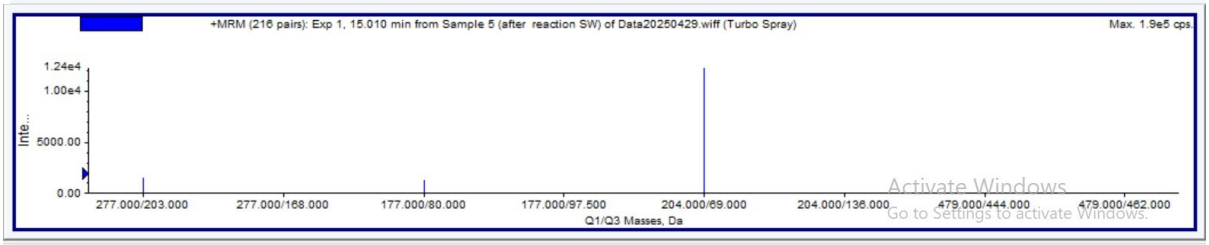

-MRM (87 pairs): Exp 2, 2.601 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 2.2e4 cps.
Inte...
7601
6000
4000
2000
0
335.000/265.000
335.000/69.000
241.000/226.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

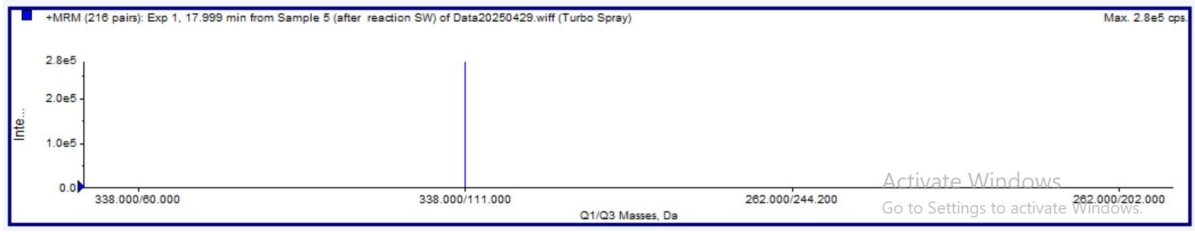

+MRM (216 pairs): Exp 1, 11.148 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.7e5 cps.
Inte...
1400
1000
500
0
195.000/44.500
237.000/194.000
237.000/192.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

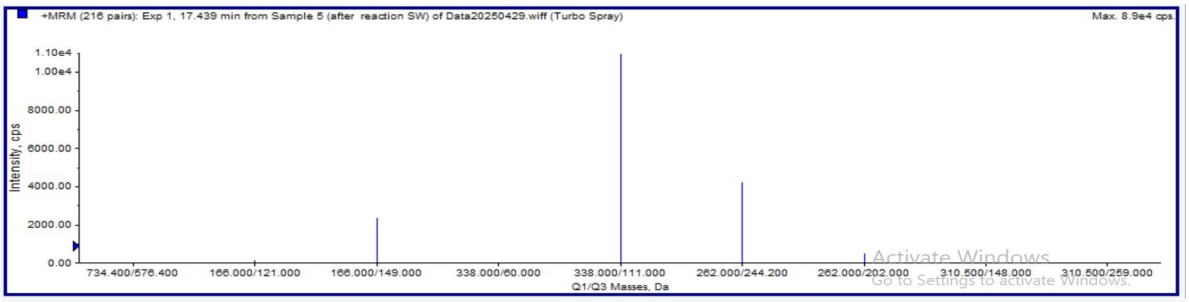

+MRM (216 pairs): Exp 1, 14.325 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 2.6e5 cps.
Inte...
2.6e5
2.0e5
1.0e5
0.0
274.100/102.000
274.100/88.000
274.100/70.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

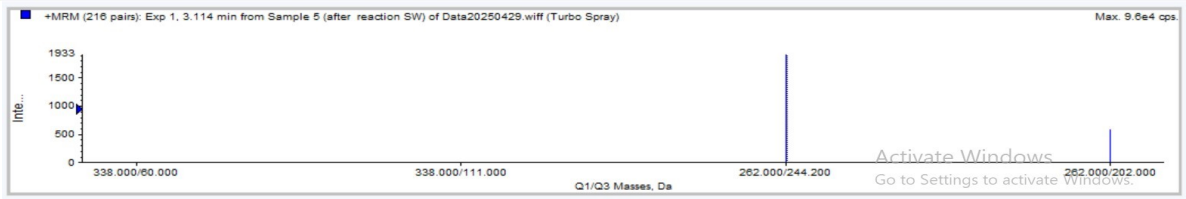

+MRM (216 pairs): Exp 1, 21.736 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.2e5 cps.
Intensity, cps
6601
6000
5000
4000
3000
2000
1000
0
748.400/590.400
277.000/203.000
277.000/168.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

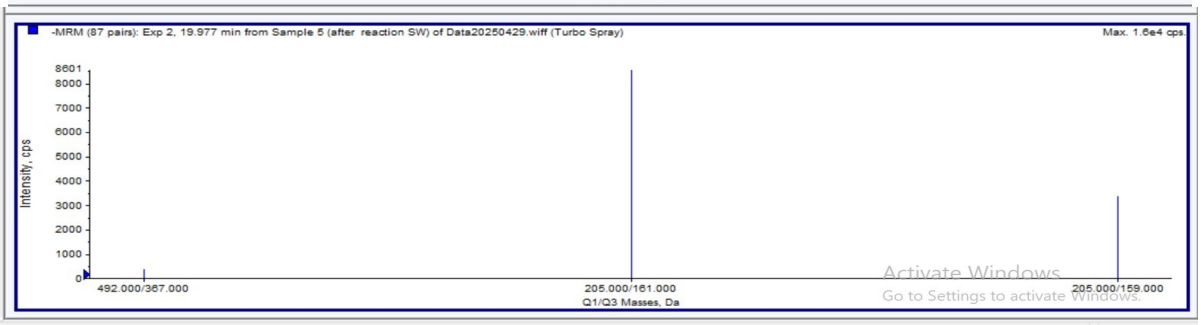

-MRM (87 pairs): Exp 2, 0.732 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 3.0e5 cps.
Inte...
3.0e5
2.0e5
1.0e5
0.0
294.200/134.200
235.100/191.000
235.100/73.100
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

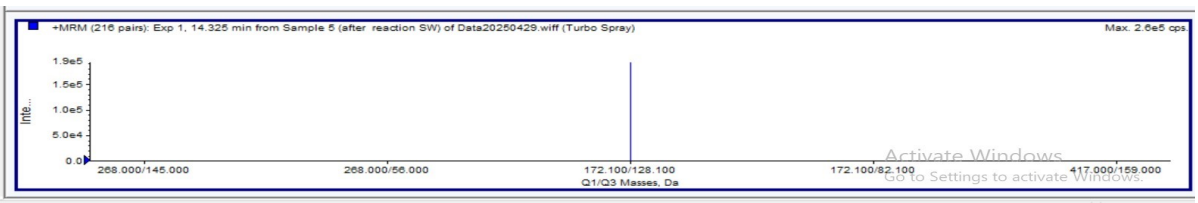

+MRM (216 pairs): Exp 1, 15.010 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.9e5 cps.
Inte...
1.24e4
1.00e4
5000.00
0.00
277.000/203.000
277.000/168.000
177.000/80.000
177.000/97.500
204.000/69.000
204.000/136.000
479.000/444.000
479.000/462.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

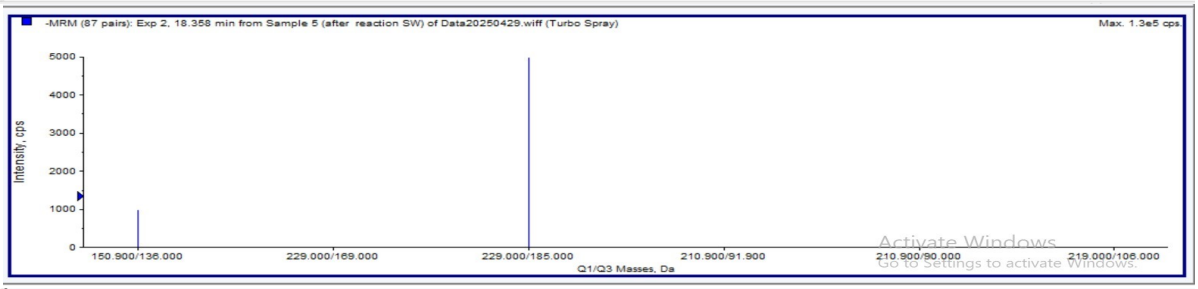

+MRM (216 pairs): Exp 1, 17.999 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 2.8e5 cps.
Inte...
2.8e5
2.0e5
1.0e5
0.0
338.000/60.000
338.000/111.000
262.000/244.200
262.000/202.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

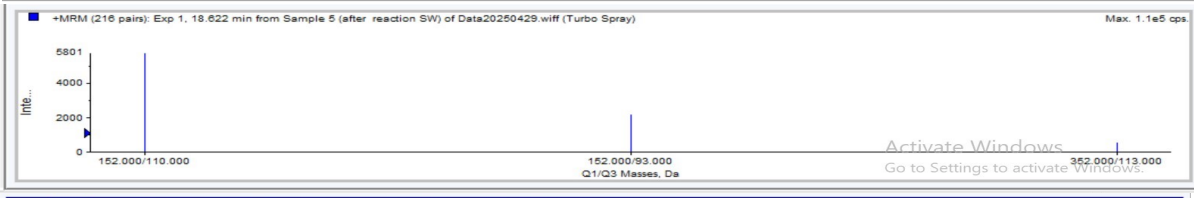

+MRM (216 pairs): Exp 1, 17.439 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 8.9e4 cps.
Intensity, cps
1.10e4
1.00e4
8000.00
6000.00
4000.00
2000.00
0.00
734.400/576.400
166.000/121.000
166.000/149.000
338.000/60.000
338.000/111.000
262.000/244.200
262.000/202.000
310.500/148.000
310.500/259.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 3.114 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 9.6e4 cps.
Inte...
1933
1500
1000
500
0
338.000/60.000
338.000/111.000
262.000/244.200
262.000/202.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


-MRM (87 pairs): Exp 2, 19.977 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.6e4 cps.
Intensity, cps
8601
8000
7000
6000
5000
4000
3000
2000
1000
0
492.000/367.000
205.000/161.000
205.000/159.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 14.325 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 2.6e5 cps.
Inte...
1.9e5
1.5e5
1.0e5
5.0e4
0.0
268.000/145.000
268.000/56.000
172.100/128.100
172.100/82.100
417.000/159.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


-MRM (87 pairs): Exp 2, 18.358 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.3e5 cps.
Intensity, cps
5000
4000
3000
2000
1000
0
150.900/136.000
229.000/169.000
229.000/185.000
210.900/91.900
210.900/90.000
219.000/106.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.


+MRM (216 pairs): Exp 1, 18.622 min from Sample 5 (after reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 1.1e5 cps.
Inte...
5801
4000
2000
0
152.000/110.000
152.000/93.000
352.000/113.000
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

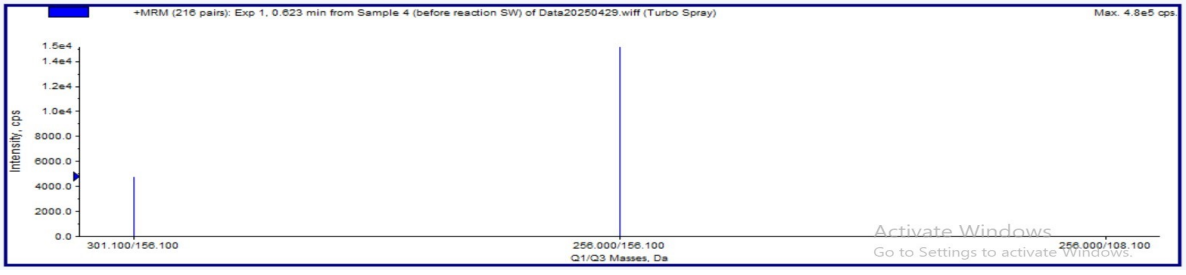
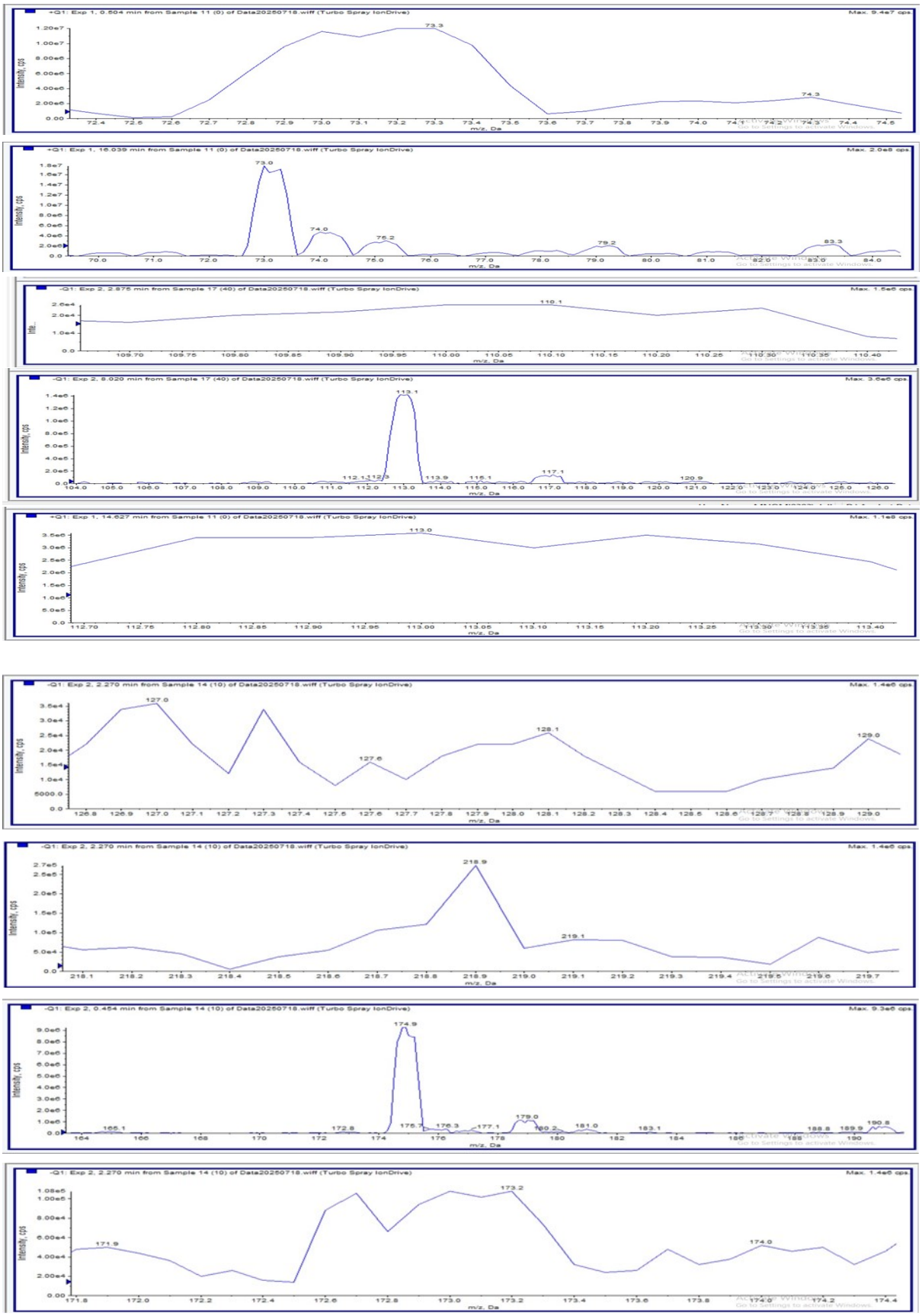
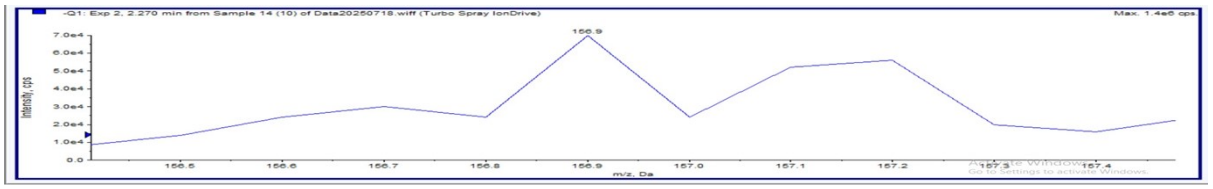
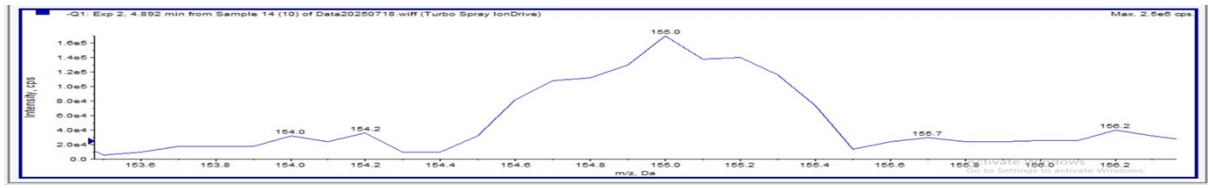
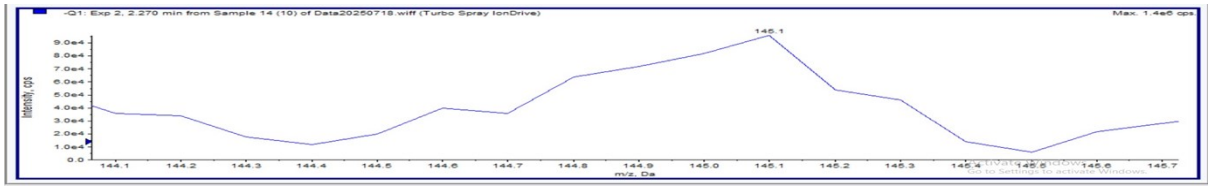

+MRM (216 pairs): Exp 1, 0.623 min from Sample 4 (before reaction SW) of Data20250429.wiff (Turbo Spray)
Max. 4.8e5 cps.
Intensity, cps
1.5e4
1.4e4
1.2e4
1.0e4
8000.0
6000.0
4000.0
2000.0
0.0
301.100/156.100
256.000/156.100
256.000/108.100
Q1/Q3 Masses, Da
Activate Windows
Go to Settings to activate Windows.

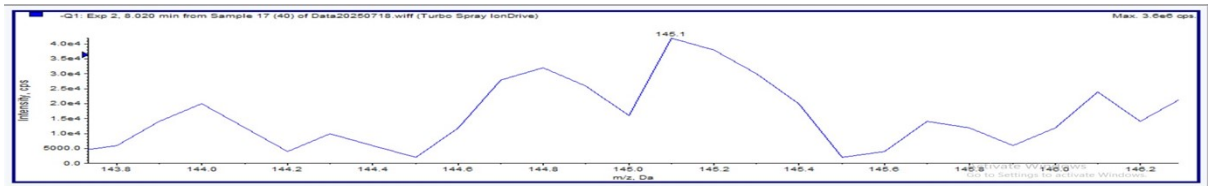
Figure S7: OTC intermediates (After reaction)

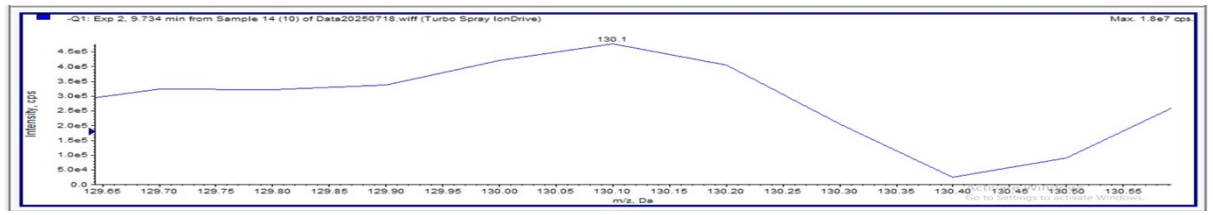


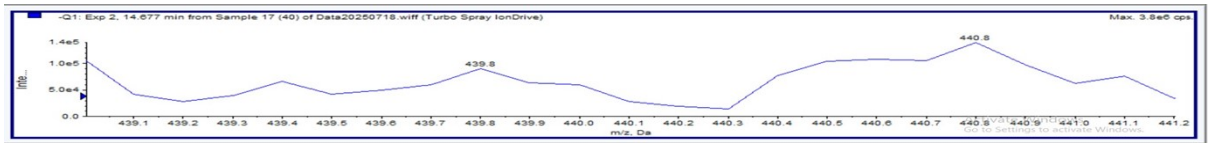

-Q1: Exp 2, 2.270 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.4e6 cps.
Intensity, cps
m/z, Da

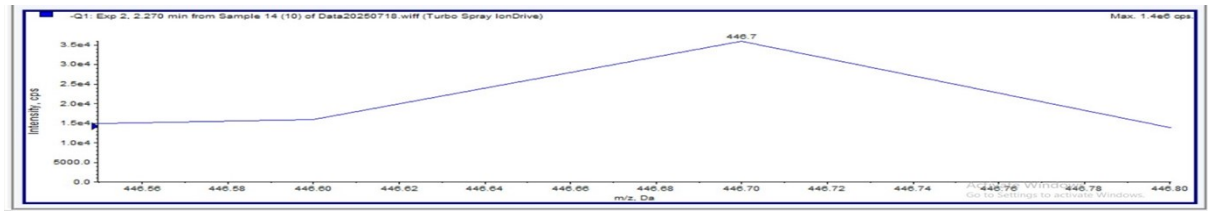

-Q1: Exp 2, 4.892 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.5e6 cps.
Intensity, cps
m/z, Da

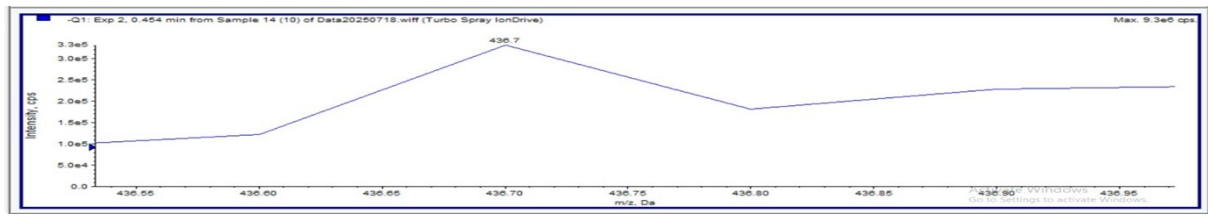

-Q1: Exp 2, 2.270 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.4e6 cps.
Intensity, cps
m/z, Da

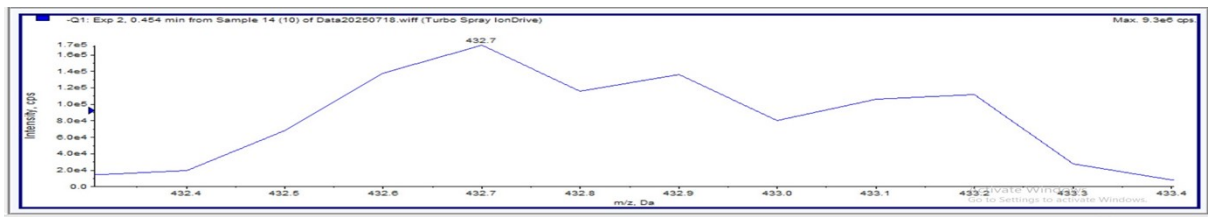

-Q1: Exp 2, 8.020 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 3.6e6 cps.
Intensity, cps
m/z, Da

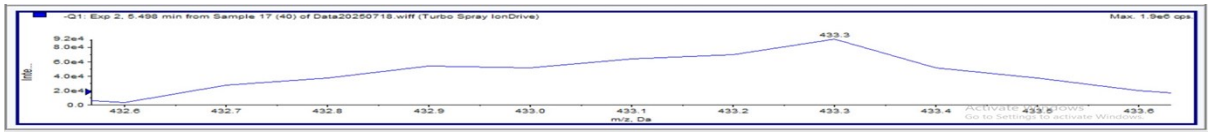

-Q1: Exp 2, 9.734 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.8e7 cps.
Intensity, cps
m/z, Da

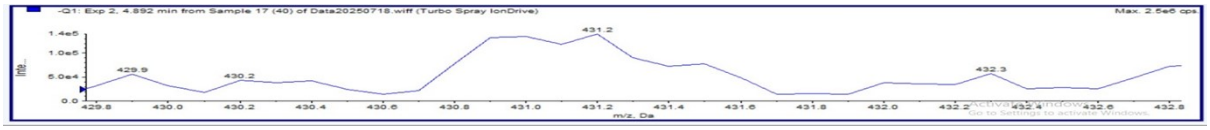

-Q1: Exp 2, 14.677 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 3.8e6 cps.
m/z, Da

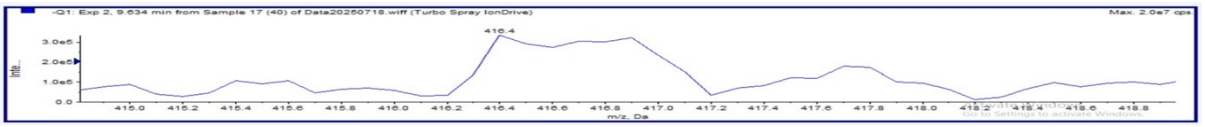

-Q1: Exp 2, 2.270 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.4e6 cps.
Intensity, cps
m/z, Da

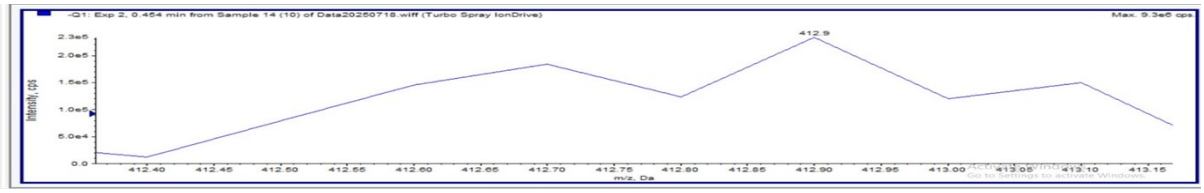

-Q1: Exp 2, 0.454 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.3e6 cps.
Intensity, cps
m/z, Da

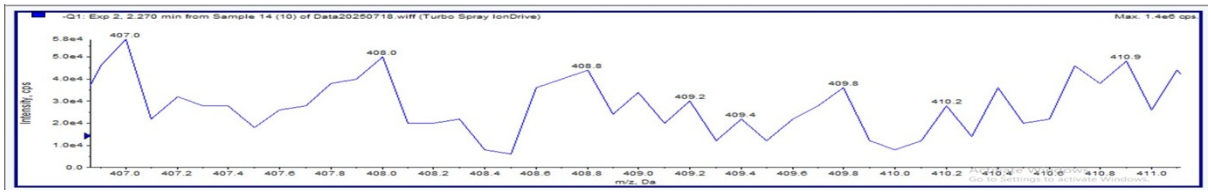

-Q1: Exp 2, 0.454 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.3e6 cps.
Intensity, cps
m/z, Da

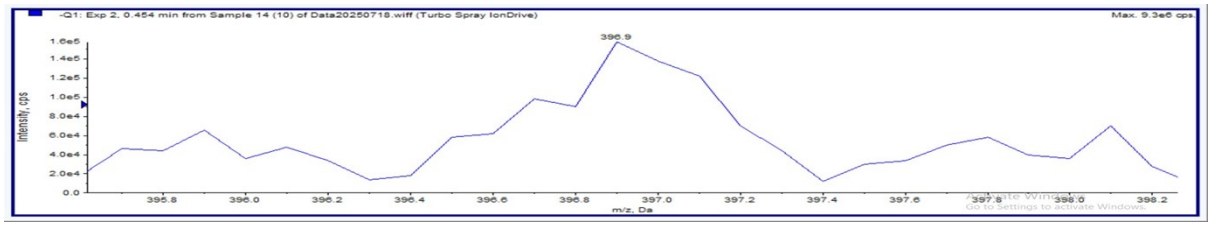

-Q1: Exp 2, 5.498 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.9e6 cps.
Inte...
433.3
m/z, Da

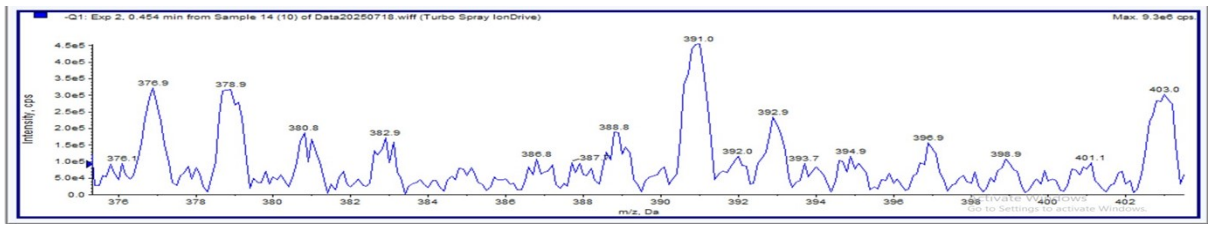

-Q1: Exp 2, 4.892 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.5e6 cps.
Inte...
429.9
430.2
431.2
432.3
m/z, Da

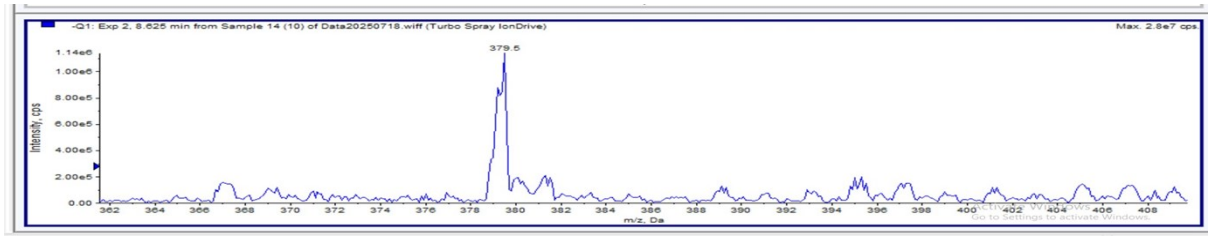

-Q1: Exp 2, 9.634 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.0e7 cps.
Inte...
416.4
m/z, Da

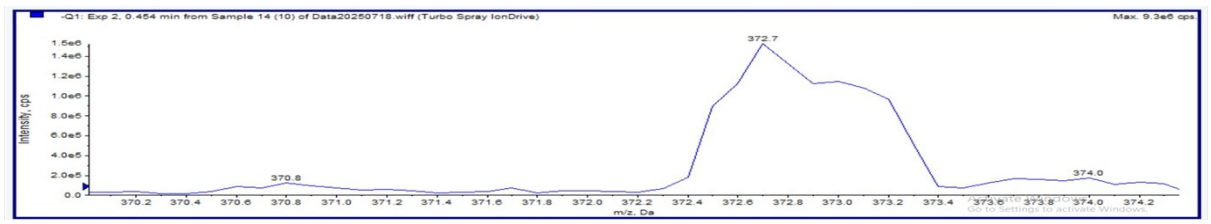

-Q1: Exp 2, 0.454 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.3e6 cps.
Intensity, cps
412.9
m/z, Da

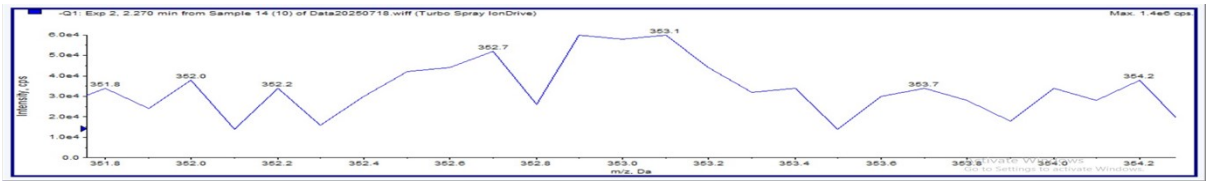

-Q1: Exp 2, 2.270 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.4e6 cps.
Intensity, cps
407.0
408.0
408.8
409.2
409.4
409.8
410.2
410.9
m/z, Da

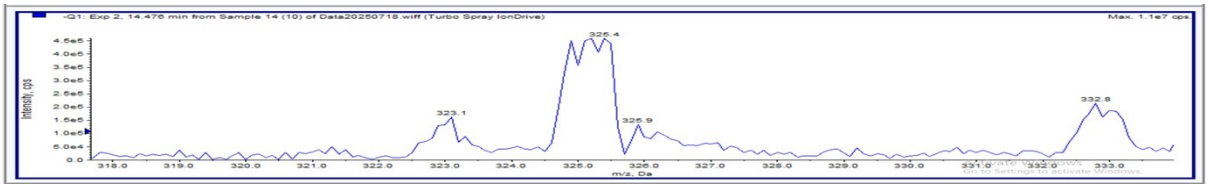

-Q1: Exp 2, 0.454 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.3e6 cps.
Intensity, cps
396.9
m/z, Da

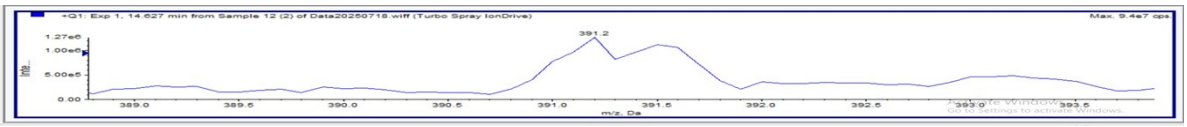

-Q1: Exp 2, 0.454 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.3e6 cps.
Intensity, cps
376.7
376.9
378.9
380.8
382.9
386.8
387.7
388.8
391.0
392.0
392.9
393.7
394.9
396.9
398.9
401.1
403.0
m/z, Da

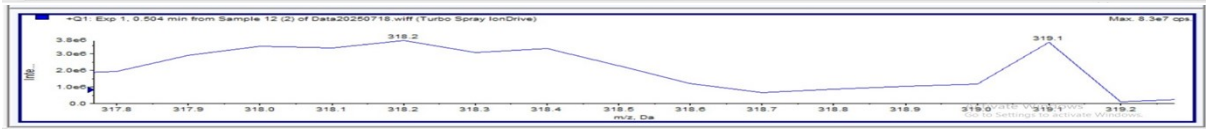

-Q1: Exp 2, 8.625 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.8e7 cps.
Intensity, cps
379.5
m/z, Da

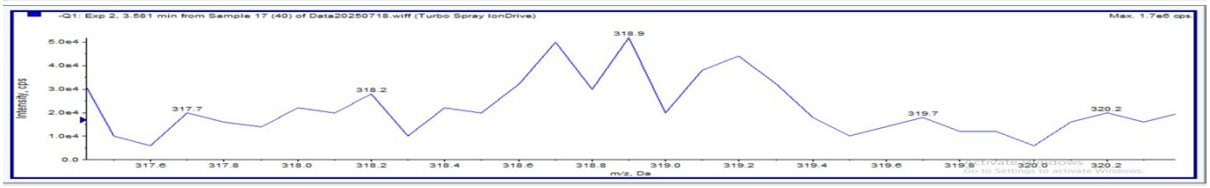

-Q1: Exp 2, 0.454 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.3e6 cps.
Intensity, cps
370.8
372.7
374.0
m/z, Da

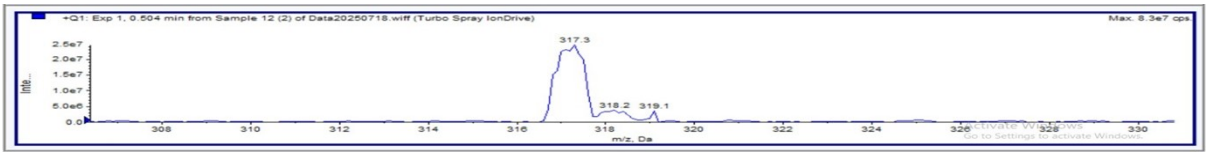

-Q1: Exp 2, 2.270 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.4e6 cps.
Intensity, cps
m/z, Da
351.8
352.0
352.2
352.7
353.1
353.7
354.2

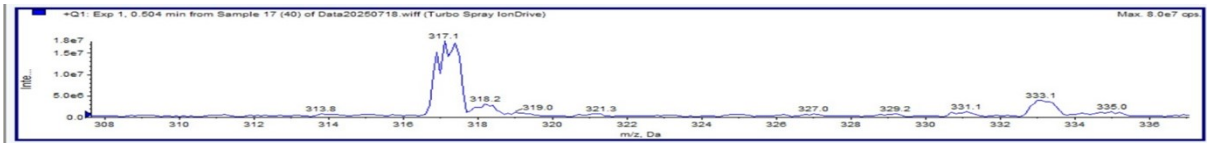

-Q1: Exp 2, 14.476 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.1e7 cps.
Intensity, cps
m/z, Da
323.1
325.4
325.9
332.8

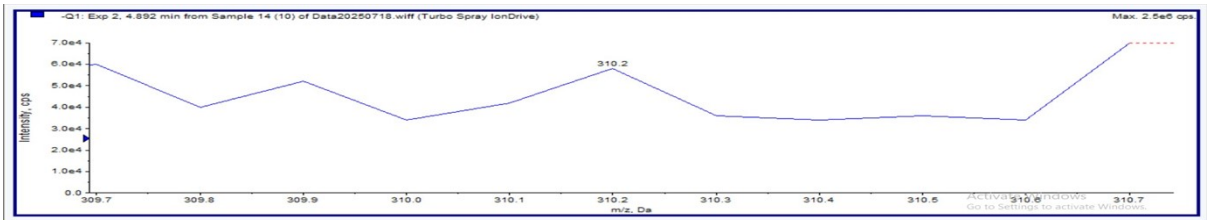

+Q1: Exp 1, 14.627 min from Sample 12 (2) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 9.4e7 cps.
m/z, Da
391.2

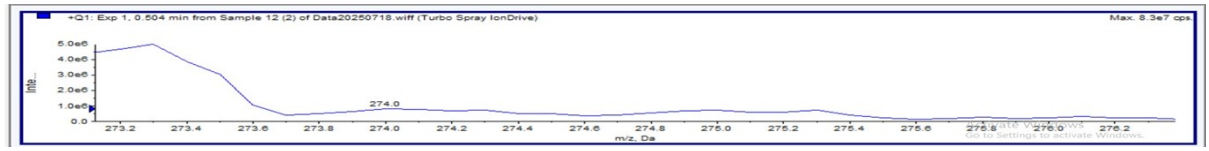

+Q1: Exp 1, 0.504 min from Sample 12 (2) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 8.3e7 cps.
m/z, Da
318.2
319.1

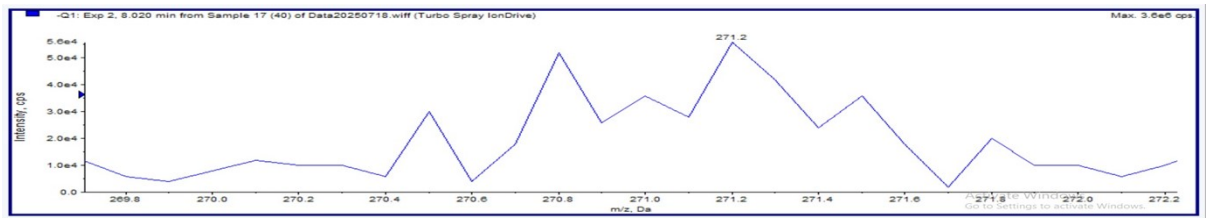

-Q1: Exp 2, 3.581 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.7e6 cps.
Intensity, cps
m/z, Da
317.7
318.2
318.9
319.7
320.2

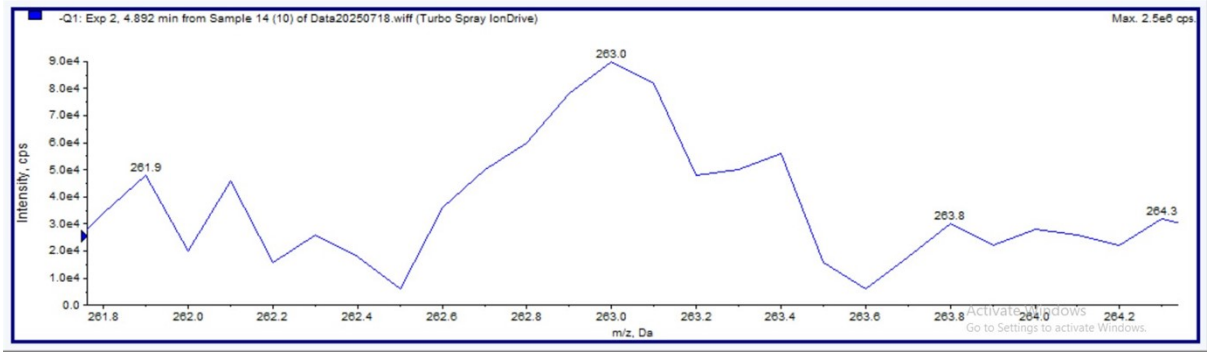

+Q1: Exp 1, 0.504 min from Sample 12 (2) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 8.3e7 cps.
m/z, Da
317.3
318.2
319.1

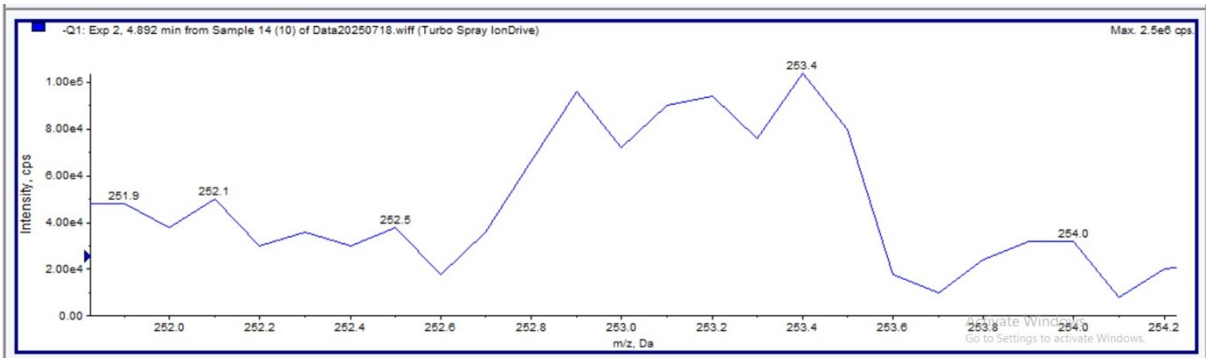

+Q1: Exp 1, 0.504 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 8.0e7 cps.
m/z, Da
313.8
317.1
318.2
319.0
321.3
327.0
329.2
331.1
333.1
335.0


-Q1: Exp 2, 4.892 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.5e6 cps.
Intensity, cps
m/z, Da
310.2


+Q1: Exp 1, 0.504 min from Sample 12 (2) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 8.3e7 cps.
m/z, Da
274.0


-Q1: Exp 2, 8.020 min from Sample 17 (40) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 3.6e6 cps.
Intensity, cps
m/z, Da
271.2


-Q1: Exp 2, 4.892 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.5e6 cps.
Intensity, cps
m/z, Da
261.9
263.0
263.8
264.3


-Q1: Exp 2, 4.892 min from Sample 14 (10) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 2.5e6 cps.
Intensity, cps
m/z, Da
251.9
252.1
252.5
253.4
254.0

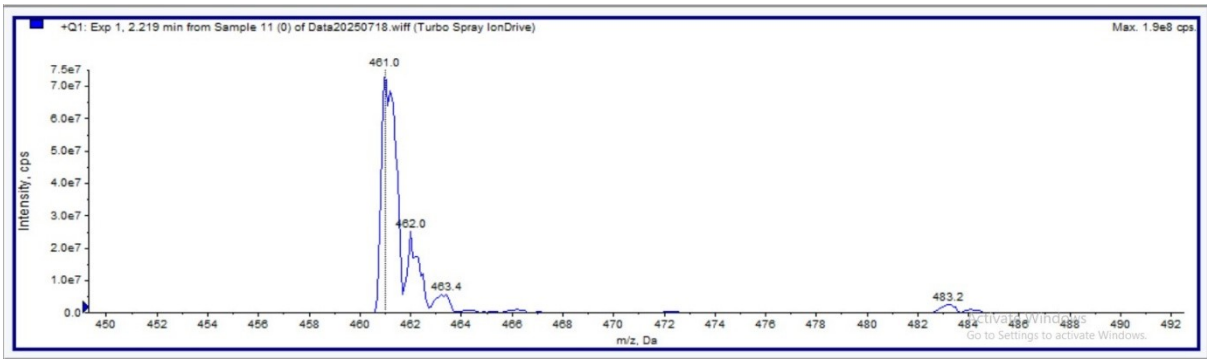

+Q1: Exp 1, 2.219 min from Sample 11 (0) of Data20250718.wiff (Turbo Spray IonDrive)
Max. 1.9e8 cps.
Intensity, cps
m/z, Da
461.0
462.0
463.4
483.2

Fig. S8: Adsorption of OTC over 2.5%Ni/Fe-C-700℃

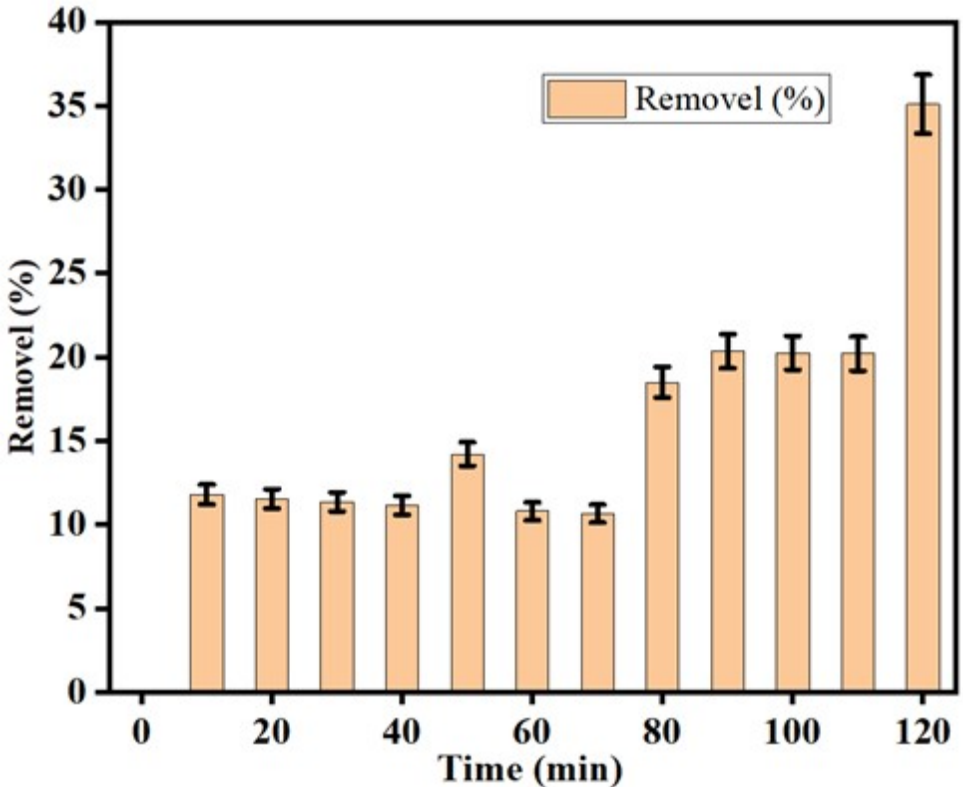


Table S1. Raw Partially reduced iron XRF results

Analyte	Calibration Status	Compound Formula	Concentration	Unit
Mg	Calibrated	MgO	1.103	%
Al	Calibrated	Al_2O_3	1.348	%
Si	Calibrated	SiO_2	16.904	%
P	Calibrated	P_2O_5	0.673	%
S	Calibrated	SO_3	0.821	%
Cl	Calibrated	Cl	0.099	%
K	Calibrated	K_2O	0.087	%
Ca	Calibrated	CaO	58.312	%
Ti	Calibrated	TiO_2	2.260	%
V	Calibrated	V_2O_5	0.000	%
Cr	Calibrated	Cr_2O_3	3.634	%
Mn	Calibrated	MnO	13.084	%
Fe	Calibrated	Fe_2O_3	0.858	%
Co	Calibrated	Co_3O_4	0.091	%
Sr	Calibrated	SrO	0.065	%
Nb	Calibrated	Nb_2O_5	0.035	%
Ba	Calibrated	BaO	0.077	%
Ce	Calibrated	CeO_2	0.550	%
Zr	**Alternative**	**ZrO_2**	**0.000**	**%**

Table S2. Real Mariculture wastewater characterization

Sr.NO	Characterizations	Before reaction	After reaction
01	pH	7.9	6.19
02	TOC	120 ppm	30 ppm
03	BOD	80 ppm	20 ppm
04	DO	5 ppm	8 ppm
05	COD		
06	Turbidity	30 NTU	5 NTU
07	Suspended solids	250 mg/L	48 mg/L
08	Ammonia nitrogen (NH3-N)	7.06 mg/L	0.86 mg/L

Table S3: Organic Pharmaceuticals Detected in Wastewater Before Treatment

Pollutant Type	Pollutant Name	Highest Intensity (cps)	Key MRM Transition (Q1/Q3)
Endocrine Disruptors	Bisphenol AF (BPAF)	2.4×10^4	335.000/265.000
Ketones	Acetophenones	1.3×10^5	268.300/152.000
Pharmaceuticals	17α-Ethinylestradiol (EE2)	3.6×10^5	293.000/221.000
	Amoxicillin	1.8×10^6	420.000/342.000
	Carbamazepine	1.8×10^3	237.000/194.000
	Ceftizoxime	6.0×10^5	460.300/361.200
	Clenbuterol	1.8×10^6	277.000/203.000
	Diclofenac	1.5×10^4	294.000/260.000
	Famotidine	3.6×10^3	166.000/121.000

Pollutant Type	Pollutant Name	Highest Intensity (cps)	Key MRM Transition (Q1/Q3)
	Fenoprofen	4.7×10^4	241.000/93.000
	Flumequine	1.5×10^3	262.200/244.000
	Ibuprofen	3.0×10^5	296.200/212.900
	Lincomycin	1.8×10^3	362.200/261.200
	Metronidazole	8.9×10^3	172.100/128.100
	Naproxen	1.3×10^5	228.000/169.000
	Paracetamol	4.7×10^4	152.000/110.000
	Sulfathiazole	4.8×10^5	266.000/156.100

Table S4: Organic Pharmaceuticals Detected in Wastewater After Catalytic Treatment

Pollutant Type	Pollutant Name	Highest Intensity (cps)	Key MRM Transition (Q1/Q3)
Endocrine Disruptors	Bisphenol AF (BPAF)	2.3×10^3	335.000/266.000
Ketones	Acetophenones	9.1×10^4	274.100/256.000
Pharmaceuticals	Amoxicillin	ND*	420.000/342.000
	Carbamazepine	1.7×10^3	237.000/194.000
	Ceftizoxime	2.0×10^5	274.100/102.000
	Clenbuterol	1.3×10^3	277.000/203.000
	Ibuprofen	1.0×10^4	296.200/212.900
	Metronidazole	8.9×10^3	172.100/128.100

Pollutant Type	Pollutant Name	Highest Intensity (cps)	Key MRM Transition (Q1/Q3)
	Naproxen	5.2×10^3	228.000/168.000
	Paracetamol	5.8×10^3	152.000/110.000
	Sulfathiazole	9.8×10^0	256.000/156.100

Table S5: Adsorption energies and adsorption distances calculation

(Ozone over 2.5%Ni/Fe-C-700℃'s adsorption models)

Models	E catalyst (kcal/mol)	ozone (kcal/mol)	E(adsorption system) (kcal/mol)	Eads (kcal/mol)	Eads (eV)	Adsorption Distance
Fe-O	143.945239	32.873527	31.284	-145.5348	-6.3105	6.235
Ni-O	143.945239	32.873527	31.293	-145.5258	-6.3101	6.576
COOH	143.945239	32.873527	31.322	-145.4968	-6.3085	6.737
OH	143.945239	32.873527	31.319	-145.4998	-6.3086	3.915
C=O	143.945239	32.873527	31.400	-145.4188	-6.3051	4.839
C≡O	143.945239	32.873527	31.294	-145.5248	-6.3101	4.889
Defects	143.945239	32.873527	31.298	-145.5208	-6.3099	3.628
Edges	143.945239	32.873527	31.237	-145.5818	-6.3134	5.461

Table S6: OTC Fukui Indices for Electrophilic Attack (Fukui (-))

Atom	Mulliken Charge	Hirshfeld Charge
O (1)	0.008	0.013
O (2)	-0.016	-0.002
O (3)	0.008	0.008

Atom	Mulliken Charge	Hirshfeld Charge
O (4)	0.026	0.020
O (5)	0.027	0.025
O (6)	0.018	0.015
O (7)	0.045	0.037
O (8)	0.004	0.007
O (9)	0.024	0.023
N (10)	0.196	0.222
N (11)	0.010	0.016
C (12)	-0.013	-0.002
C (13)	-0.003	0.002
C (14)	-0.006	0.003
C (15)	-0.016	-0.004
C (16)	-0.046	0.022
C (17)	-0.007	0.001
C (18)	-0.005	-0.005
C (19)	-0.006	-0.004
C (20)	0.016	0.009
C (21)	-0.003	-0.002
C (22)	-0.014	-0.001
C (23)	0.011	0.011

Atom	Mulliken Charge	Hirshfeld Charge
C (24)	-0.001	0.002
C (25)	-0.003	0.004
C (26)	-0.005	-0.003
C (27)	0.002	0.006
C (28)	-0.049	0.040
C (29)	-0.049	0.042
C (30)	0.005	0.006
C (31)	0.007	0.003
C (32)	0.007	0.015
C (33)	0.005	0.011
H (34)	0.017	0.005
H (35)	0.009	0.002
H (36)	0.016	0.005
H (37)	0.090	0.051
H (38)	0.007	0.004
H (39)	0.018	0.010
H (40)	0.009	0.005
H (41)	0.011	0.008
H (42)	0.021	0.013
H (43)	0.006	0.003

Atom	Mulliken Charge	Hirshfeld Charge
H (44)	0.013	0.006
H (45)	0.072	0.038
H (46)	0.112	0.069
H (47)	0.066	0.033
H (48)	0.113	0.071
H (49)	0.071	0.041
H (50)	0.069	0.035
H (51)	0.012	0.011
H (52)	0.005	0.004
H (53)	0.020	0.010
H (54)	0.019	0.009
H (55)	0.009	0.007
H (56)	0.025	0.015
H (57)	0.010	0.007

Table S7: OTC Fukui Indices for Nucleophilic Attack (Fukui (+))

Atom	Mulliken Charge	Hirshfeld Charge
O (1)	0.014	0.016
O (2)	0.028	0.027
O (3)	0.015	0.015
O (4)	0.118	0.111

Atom	Mulliken Charge	Hirshfeld Charge
O (5)	0.029	0.030
O (6)	0.041	0.034
O (7)	0.065	0.059
O (8)	0.010	0.016
O (9)	0.042	0.036
N (10)	-0.010	-0.001
N (11)	0.008	0.012
C (12)	-0.009	0.003
C (13)	-0.012	0.004
C (14)	0.004	-0.001
C (15)	-0.018	0.014
C (16)	-0.004	0.004
C (17)	-0.008	0.003
C (18)	-0.007	0.008
C (19)	0.106	0.104
C (20)	0.024	0.024
C (21)	0.013	0.012
C (22)	0.019	0.026
C (23)	0.036	0.040
C (24)	0.031	0.032

Atom	Mulliken Charge	Hirshfeld Charge
C (25)	-0.007	0.005
C (26)	-0.007	0.003
C (27)	0.007	0.019
C (28)	-0.013	0.005
C (29)	-0.017	0.005
C (30)	0.019	0.017
C (31)	0.004	0.006
C (32)	0.023	0.037
C (33)	0.010	0.022
H (34)	0.025	0.010
H (35)	0.023	0.010
H (36)	0.008	0.003
H (37)	0.027	0.014

H37	0.027	0.014
H38	0.021	0.010
H39	0.025	0.013
H40	0.004	0.003
H41	0.014	0.010
H42	0.019	0.013
H43	0.014	0.010

Atom	Mulliken Charge	Hirshfeld Charge
H44	0.030	0.015
H45	-0.001	0.001
H46	0.020	0.010
H47	0.027	0.014
H48	0.021	0.010
H49	0.004	0.004
H50	0.027	0.014
H51	0.012	0.011
H52	0.008	0.012
H53	0.039	0.021
H54	0.035	0.017
H55	-0.004	0.001
H56	0.028	0.016
H57	0.019	0.015

Table S8: OTC Fukui Indices for Radical attack (Fukui (0))

Atom	Mulliken Charge	Hirshfeld Charge
O (1)	0.011	0.015
O (2)	0.006	0.012
O (3)	0.011	0.012
O (4)	0.072	0.065

Atom	Mulliken Charge	Hirshfeld Charge
O (5)	0.028	0.028
O (6)	0.029	0.025
O (7)	0.055	0.048
O (8)	0.007	0.011
O (9)	0.033	0.030
N (10)	0.093	0.111
N (11)	0.009	0.014
C (12)	-0.011	0.000
C (13)	-0.007	0.003
C (14)	-0.001	0.001
C (15)	-0.017	0.005
C (16)	-0.025	0.013
C (17)	-0.008	0.002
C (18)	-0.006	0.002
C (19)	0.050	0.050
C (20)	0.020	0.016
C (21)	0.005	0.005
C (22)	0.003	0.012
C (23)	0.024	0.025
C (24)	0.015	0.017

Atom	Mulliken Charge	Hirshfeld Charge
C (25)	-0.005	0.005
C (26)	-0.006	0.000
C (27)	0.005	0.013
C (28)	-0.031	0.022
C (29)	-0.033	0.023
C (30)	0.012	0.011
C (31)	0.005	0.004
C (32)	0.015	0.026
C (33)	0.008	0.016
H (34)	0.021	0.008
H (35)	0.016	0.006
H (36)	0.012	0.004
H (37)	0.059	0.032
H (38)	0.014	0.007
H (39)	0.022	0.011
H40	0.006	0.004
H41	0.012	0.009
H42	0.020	0.013
H43	0.010	0.006
H44	0.021	0.010

Atom	Mulliken Charge	Hirshfeld Charge
H45	0.036	0.019
H46	0.066	0.039
H47	0.046	0.023
H48	0.067	0.040
H49	0.038	0.022
H50	0.048	0.024
H51	0.012	0.011
H52	0.006	0.008
H53	0.030	0.016
H54	0.027	0.013
H55	0.002	0.004
H56	0.026	0.016
H57	0.015	0.011

Table S9: OTC degradation efficiency with other studies.

Number of Studies	Pollutant load	Catalyst dosage	Removal rate (%)	Ozone concentration/PMS	Refferenes
This work	10 mg/L	0.1 g/L	96.7	6.5	This work
1	20 mg/L	0.2 g/L	100	0.2mM	[9]
2	20 mg/L	0.4 g/L	92.81	Adsorption	[10]
3	10.68 mg/L	0.46 g/L	99.52	0.21 g/L	[11]
4	270 mg/L	0.5 g/L	89.5	10 mg/L	[12]
5	5 mg/L	0.25 g/L	89.5	adsorption	[13]
6	10 mg/L	0.25 g/L	84.5	Photocatalytic reaction	[14]
7	10 mg/L	1 g/L	85.9	80 mM (PMS)	[15]
8	20 mg/L	0.20g/100mL	94.38	Photocatalytic reaction	[16]
9	10 mg/L	0.1 g/L	99.9	1 mM (PMS)	[17]

10	5 mg/L	0.2 g/L	97.9	Photocatalytic reaction	[18]
11	50 mM	0.1 g/L	90.92	5 mM	[19]
12	30 mg/L	0.01 mole/L	96	Photo-Fenton degradation	[20]

Table S10: Life-cycle inventory (LCI) based attributional cradle-to-grave LCA calculations

Metric	**Result per 0.5 kg catalyst**	**Greener ?**	**Why It's Greener**
Avoided Burden - Coconut Shell	0.30 kg CO_2-eq saved	Yes	Diverts agricultural waste from landfill, reducing methane emissions
Avoided Burden - Iron Fines	0.01 kg CO_2-eq saved	Yes	Utilizes industrial by-products that would otherwise go to waste
Toxicity Impact (Ni)	1.55×10^{-6} CTUh	Yes	Nickel toxicity impact is negligible (extremely low)
Circularity (4 cycles)	4.4% circular	Yes	Enables multiple reuse cycles, reducing virgin material demand
Regeneration Credit	3.27 kg CO_2-eq saved	Yes	Regeneration uses 81% less CO_2 than making a new catalyst
Net Environmental Benefit	**3.58 kg CO_2-eq net saved**	Yes	**Overall significant carbon reduction + waste valorization**

Life-cycle inventory (LCI) based attributional cradle-to-grave LCA

1 Avoided-burden credit for waste utilization

1. 2 Coconut Shell Residue (Pyrolysed to Biochar)

- **Mass (m_CNR):** 0.30 kg (60% of the 0.5 kg composite)
- **Disposal CF (Landfill, GWP):** 1.0 kg CO_2-eq kg^{-1} (for wet biomass, IPCC 2019)
- **Avoided GWP Impact:**

0.30 kg × 1.0 kg CO2 - eq kg - 1 = 0.30 kg CO_2 - eq

1.3 Partially-Reduced Iron-Ore Fines (RI)

- **Mass (m_RI):** 0.20 kg (40% of the 0.5 kg composite)

- **Disposal CF (Landfill, GWP):** 0.05 kg CO_2-eq kg^{-1} (for inert mineral waste)
- **Avoided GWP Impact:**

$0.20\,kg \times 0.05\,kg\ CO_2 - eq\ kg - 1 = \ 0.01\,kg\ CO_2 - eq$

2. Toxicity increment from impurities in waste feedstock (Based on EDS)

EDS Analysis of 0.5 kg Catalyst:

- Nickel (Ni): 2.38% = 0.0119 kg
- Iron (Fe): 0.27% = 0.00135 kg
- Carbon (C): 6.96% = 0.0348 kg (non-toxic)
- Oxygen (O): 36.14% = 0.1807 kg (non-toxic)

USEtox Factors (Human Toxicity, Soil Emission):

- Ni: 1.3×10^{-4} CTUh kg^{-1}
- Fe: Considered non-toxic in LCA (CF ≈ 0)

Toxicity Calculation:

- Ni Impact = 0.0119 kg × 1.3×10^{-4} CTUh kg^{-1} = **1.55×10^{-6} CTUh**
- Fe Impact = 0.00135 kg × 0 CTUh kg^{-1} = **0 CTUh**
- **Total Impact = 1.55×10^{-6} CTUh**

Why Only Ni Matters:

- Ni has a high concentration (2.38%) and significant toxicity
- Fe is an essential nutrient, non-toxic in LCA models
- C and O are non-toxic elements
- No other metals detected in EDS analysis

Final: Toxicity Increment = 1.55×10^{-6} CTUh per m^3 wastewater.

3. Resource Efficiency & Circularity Metrics (Catalyst Life Cycle)

Basic Formula:

MCI = Waste fraction × (Waste fraction) ^ (cycles-1)

Where:

- Waste fraction = 45.75% = 0.4575
- Cycles = 4

Calculation:

MCI = $0.4575 \times (0.4575)^3$

= 0.4575 × 0.4575 × 0.4575 × 0.4575

= **0.044 (or 4.4%)**

Cycle-by-cycle breakdown:

- After 1 cycle: 45.75% circular
- After 2 cycles: 20.9% circular
- After 3 cycles: 9.6% circular
- After 4 cycles: **4.4% circular**

4. Regeneration Efficiency and Impact (Catalyst Discharge/End-of-Life)

Given Data:

- Fresh catalyst impact: 2.1 kg CO_2-eq/kg
- Regenerated catalyst impact: 0.4 kg CO_2-eq/kg
- Cycles: 5

- Recovery efficiency: 95% per cycle

Calculation Formula:

Credit = (Impact_fresh - Impact_reg) × Effective Cycles

Step 1: Calculate Savings Per Cycle

2.1 - 0.4 = 1.7 kg CO_2-eq/kg per regeneration

Step 2: Calculate Effective Cycles (Accounting for 5% Loss)

After 5 cycles: $0.95^5 = 0.77$ (77% mass remains)

Effective cycles = 5 × 0.77 = 3.85 cycles

Step 3: Calculate Total Credit

Credit = 1.7 × 3.85 = 6.55 kg CO_2-eq/kg catalyst

Step 4: Apply to Functional Unit (0.5 kg catalyst)

6.55 × 0.5 = 3.27 kg CO_2-eq saved

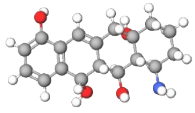
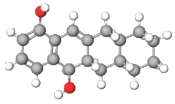
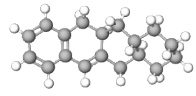
Final Result:

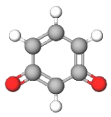
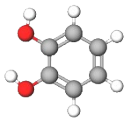
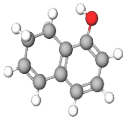
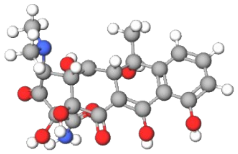
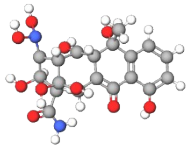
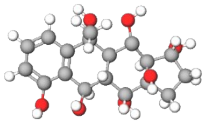
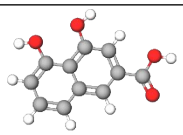
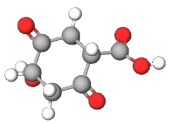
3.27 kg CO_2-eq saved per 0.5 kg catalyst over 5 regeneration cycles.

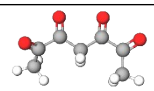
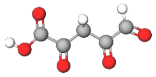
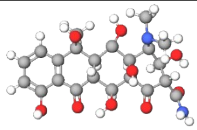
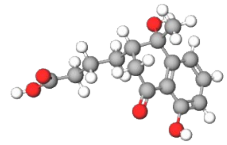
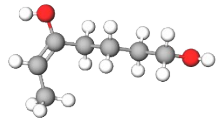
Table S11: Comprehensive comparison of 2.5%Ni/Fe-C-700°C with other catalysts based on other factors

Sr. No	Material	OTC concentration	Cost estimation	Removal efficiency	Ozone consumption	References
01	Z5Å-Co-Fe/O_3	OTC in a real matrix	Not report	95 % (15 min, pH 6)	1.6 mg min^{-1}	[21]
02	Mn1-Mg2/Al2O3	OTC hydrochloride	not reported	82.5–89.5 % OTC,	not reported	[12]
03	Mn1-Mg2/Al2O3	OTC hydrochloride	not reported	82.5–89.5 % OTC	not reported	[12]
04	CaO_2/O_3	WWTP sludge	not reported	90 % VS,	CaO_2/O_3	[22]
05	Bi_2S_3@NH_2-MIL-125(Ti)	10 mg/L tocilizumab		176 % TOC (15 min)	5 L h^{-1}	[11]
07	SCBW-10 (SCB/WS_2)	5 mg L^{-1} OTC	not reported	92.7 % (180 min, pH 7)	none (photocatalysis)	[23]
09	*pm*SA800	20 mg/L	1.60 USD	0.2mM	20 mg/L	[23]
08	**2.5%Ni/Fe-C-700°C**	**10 mg/L**	**1.547 USD (waste-based)**	**96.74%**	**5.5± 0.5 mg/L**	**This study**

Table S12: Toxicity assessment of all intermediates of OTC over 2.5%Ni/Fe-C-700°C/O_3

Sr. No	Intermediates	Bioconcentration_	Daphnia magna LC50_(48_hr)	Developmental	Mutagenicity
OTC		2.78	11.98	Developmental toxicant	Mutagenicity Positive
P1		2.78	11.98	Developmental toxicant	Mutagenicity Positive
P2		N/A	N/A	N/A	N/A
P3		N/A	N/A	N/A	N/A
P4		7.34	3.25	Developmental toxicant	Mutagenicity Negative
P5		76.81	2.62	Developmental toxicant	Mutagenicity Positive
P6		810.47	0.20	Developmental toxicant	Mutagenicity Positive

P7		**3.20**	**227.64**	**Developmental toxicant**	**Mutagenicity Negative**
P8		**5.05**	**11.58**	**Developmental toxicant**	**Mutagenicity Negative**
P9		**17.25**	**5.39**	**Developmental toxicant**	**Mutagenicity Positive**
R1		**N/A**	**N/A**	**N/A**	**N/A**
R2		**N/A**	**N/A**	**Developmental toxicant**	**Mutagenicity Positive**
R3		**2.49**	**72.85**	**Developmental toxicant**	**Mutagenicity Negative**
R4		**17.58**	**4.44**	**Developmental toxicant**	**Mutagenicity Negative**
R5		**5.91E-02**	**681.86**	**Developmental NON-toxicant**	**Mutagenicity Negative**

R6a		**0.19**	**218.03**	**Developmental toxicant**	**Mutagenicity Negative**
R6b		**3.59E-02**	**274.64**	**Developmental toxicant**	**Mutagenicity Negative**
T1		**4.60**	**68.65**	**Developmental toxicant**	**Mutagenicity Positive**
T2		**N/A**	**N/A**	**Developmental toxicant**	**N/A**
T3		**2.30**	**186.00**	**Developmental toxicant**	**Mutagenicity Negative**

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