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Comments on the reduction in emerging contaminants in water samples from the Esmeraldas coast (Ecuador)

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Dear Editor,

We read with great interest the recent article by Cipriani-Avila et al., where the authors showed that pharmacological micropollutants (indicated as emerging contaminants, ECs), reported in water samples of Esmeraldas Coast (Ecuador) throughout 10 river mouth sites and 14 coastal sites, exhibited a marked difference in concentration between two different annual periods, one of which characterized by COVID-19-caused social restrictions and lockdowns (Cipriani-Avila et al., 2023). Briefly speaking, the authors evaluated the concentration in the indicated samples of at least four commonly used pharmaceutical drugs (acetaminophen, diclofenac, sulphomethoxazole and trimethoprim) and of caffeine, in two different experimental settings, i.e., in a COVID-19-free period (November 2019) and during the COVID-19 pandemic (November 2020), in order to use drugs as ECs of anthropic presence. I found the idea to assess the dynamic of COVID-19 pandemic by detecting ECs in the environment very intriguing, yet some questionable issues were raised when I thoroughly read the paper.

The analytical chemistry of ECs determination (Cipriani-Avila et al., 2023), appeared to account only on the native bioactive molecule in the environment, not to any further degraded by-product from the same molecule due to organic or biological catabolism. This may be fundamental to ascertain microbiological activity on those molecules within the tested environment. The authors reported that environmental parameters in river mouths and coastal sites, particularly for temperature, differed significantly. Moreover, Cipriani-Avila et al., showed that there was a clear spatial relationship in the concentrations of these micropollutants in different coastal sites, as all the pharmaceutical drugs were detected in at least two of the fours coastal sites investigated and all reached a drastic reduction during the COVID-19 period (Cipriani-Avila et al., 2023).

I read with great interest this article, which raised some fundamental questions I would like to discuss in the following bullet points.

First, the authors evaluated the possibility that, due to the numerous social restrictions and lockdowns, the reduction in ECs in water may be simply explained as caused by a significant reduction in the local anthropic presence, i.e., people attending those places, particularly if tourists.

Second, despite the authors collected their specimens in the same seasonal context, environmental differences were reported, particularly for temperature. Sampling was performed via grab samples, whereas more representative samples, taken over a longer period of time, were not considered. The paper would gain great significance if the latter issue was considered. Differences in the water temperature are a possible marker of a different biological (for example microbiological) activity (Stres et al., 2008). Moreover, I wondered why pharmaceutical drugs, particularly acetaminophen, were reduced just during their highest use, namely upon the COVID-19 pandemic, if resident population may have used it even during the indicated period.

Investigating the microbiological component would provide additional insights into the problem evaluation. Unfortunately, Cipriani-Avila et al., showed additional conceptual drawbacks to fully comprehend the manuscript, probably reporting too insufficient data to conclude that reduced concentrations of selected ECs were due to the active lockdown. Some of these drawbacks are mostly related to the sampling strategy.

Sampling time, either daily or monthly, may also affect sample composition (moon position affects ocean coastline variability, due to high tide-low tide movements, as and with these ones also, the currents are associated). As the authors only reported that samples were taken in November 2019 and November 2020, probably this needs more information to be provided. Both viral and microbial communities are largely affected by moon tide cycles (Chen et al., 2019).

Alternative explanations can be forwarded, anyway.

The authors should have assessed about the Gram-positive bacterial presence in the marine areas closest to the coast, as those bacteria are able to metabolize some pharmaceuticals, such as acetaminophen (Palma et al., 2021). Despite it has been estimated that at least 95 % of seawater bacteria are represented by Gram-negative species, many Gram-positive strains are retrievable in the near-coastal areas of seawater (Jensen and Fenical, 1995).

Furthermore, the authors did not detail how they selected emerging contaminants (ECs). For example, they missed carbamazepine, besides diclofenac, a known biodegradation-resistant indicator of anthropogenic pollution (Hai et al., 2018).

Gram-positive species such as *Brevibacterium frigoritolerans*, *Corynebacterium nuruki* and *Enterococcus faecium*, are able to remove about 97 ± 4 %, 97 ± 6 % and 86.9 ± 0.8 % of paracetamol (acetaminophen) at 200 mg/L, respectively (Palma et al., 2021). *B. frigotolerans* is a strictly aerobic chemo-organotrophic bacterium, usually found in environmental contexts, such as seawater, where this species interacts with Cnidaria (*Aurelia aurita)* and Ctenophora (*Mnemiopsis leidyi*) (Weiland-Bräuer et al., 2020), whereas *Enterococcus faecium* still represents a great concern for faecal contamination of anthropized coastal areas (Rebelo et al., 2021).

The possible existence of Gram-positive bacterial species in the water micro-environments, able to metabolize the chemical backbones of pharmaceutical ECs, would support the hypothesis, though apparently speculative, that the decrease in micropollutants derived from the human use of therapeutic drugs might come from the active participation of polluting micro-organisms in degrading drugs, where changes in temperature may be referred as a possible environmental marker of this activity.

Endowing the research paper with the aforementioned microbiological and environmental issues should provide Cipriani-Avila et al.'s paper with more insights about their valuable research.

The relationship of *Enterococcus faecium* with faecal contamination can support the thesis that Ecuador rivers may be possible sources of this

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Gram-positive cocci, as recent research has reported that coliforms are above the maximal threshold established by USA, European and Brazilian guidelines (Vinueza et al., 2021). Moreover, it is possible that other bacterial species, such as *Rhodococcus ruber*, which easily grows on plastic elastomers (usually polluting seawater and shores), participated in degrading pharmaceuticals available as ECs, for example diclofenac (Mor and Sivan, 2008; Ivshina et al., 2019).

Probably, the reduction in ECs interpreted as due to a reduction in living people, attending those places, should also account on the participation of degrading microbiota in waters, which may have exerted a major role.

The degree of effective lockdown in Ecuador needs to be specified for the times of sampling in order to achieve more sound and reliable evidence, if Ecuador's response to the COVID-19 pandemic started on 26 February 2020 (Molina et al., 2021). Very likely, initial sampling (November 2019) should correspond to pre-lockdown. However, the question is what level of lockdown was active in Ecuador in November 2020, when the second sampling was performed. April/May 2020 would be a more appropriate selection.

More detailed insights may highlight the evidence reported by the authors and give fundamental data about how outdoor environments changed upon COVID-19-induced lockdowns and social restrictions.

CRediT authorship contribution statement

Conceptualization; Data curation; Formal analysis; Funding acquisition; Investigation; Methodology; Project administration; Resources; Software; Supervision; Validation; Visualization; Roles/Writing - original draft; Writing - review & editing.

Data availability

No data was used for the research described in the article.

Declaration of competing interest

The Author states he has no conflict of interest.

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