



MONITOOL

new tools for water quality monitoring



DGT: EQS adaptation for nickel, cadmium and lead - Additionnal document MONITOOL extension -

June 2023
No. version: 1



Report/Deliverable by

Isabelle Amouroux
Jean-Louis Gonzalez
Stéphane Guesdon

Disclaimer

This project (n° contract: EAPA_565/2016) is co-financed by the European Regional Development Fund through the Interreg Atlantic Area Programme. The present work reflects only the author's view and the funding Programme cannot be held responsible for any use that may be made of the information it contains.

Table of Contents

1. Introduction	4
Scope	5
2. Methodology for using DGT results towards EQS_{marine water}	5
3. Determination of EQS_{DGT}	9
3.1. MONITOOL original model	9
3.2. Update of the models considering additionnal campaign 2022 data	10
3.3. Use additionnal campaign 2022 data to verify original models	11
5. Main conclusion	12
Reference.....	13

Glossary

AA-EQS: Annual Average Environmental Quality Standard

Cd: Cadmium

DGT: Diffusive Gradient in Thin film

EQS: Environmental Quality Standard

IP: Prediction interval

ICP-MS: Inductively coupled plasma mass spectrometry

LOQ: Limit of quantification

MAC-EQS: Maximum Allowable Concentration Environmental Quality Standard

Ni: Nickel

Pb: Lead

WFD: Water Framework Directive

1. Introduction

According to the Water Framework Directive (WFD), (Directive 2000/60/EC) (1) the Chemical Status of water bodies is assessed on the basis of the compliance for each priority substance of the measured concentration with Environmental Quality Standards (EQS). Directive 2008/105/EC (2) amended by Directive 2013/39/EU (3), established the maximum allowable concentrations (MAC-EQS) and annual average concentrations (AA-EQS) of four metals. Only mercury (Hg) has an AA-EQS that refers to biota (fish); the three others – lead (Pb), cadmium (Cd) and nickel (Ni) – have an EQS applicable to surface waters. In the case of metals in transitional and coastal water bodies, the EQS refers to the dissolved concentration, i.e. the dissolved phase of a water sample obtained by filtration through a 0.45 µm filter or any equivalent pre-treatment.

In aquatic environments, most chemical contaminants (metallic or organic) are present at trace levels, complicating the detection (and quantification) of contaminants by standardized analytical techniques. Additionally, in seawater, due to the saline matrix, using “conventional” techniques requires the implementation of “ultra-clean” sampling of high volumes of water and complex concentration/extraction and analytical steps. Beyond that, the low spatial and temporal representativeness of the results of spot sampling, especially in highly dynamic systems, has been pointed out, therefore the use of integrative matrix, such as molluscs or sediments, is preferred to monitor the chemical contamination of water bodies (OSPAR (4,5), French Chemical Contamination Monitoring Network in marine environment: ROCCH). The use of passive samplers, enabling low disturbance *in situ* sampling of trace element species and providing time-averaged concentration of contaminants during the deployment time, appears as a more reliable approach for the environment chemical monitoring (6). Since one of the primary objectives of the WFD is the assessment of the annual average concentrations of pollutants in waterbodies, the determination of time-integrated concentrations using passive samplers is a promising approach, which is less influenced by short-term fluctuations in concentrations than grab sampling (i.e., discrete spot sampling). The MONITOOL project focuses on the diffusive gradients in thin films (DGT) technique, which stands out as one of the most widely used passive sampler for metals (6). These samplers were developed, in the early 90s, by Lancaster University (7) (8), in collaboration with the UK Environment Agency.

Despite the wide application of DGTs for *in situ* measurements of metal speciation in numerous scientific studies (9), the use of DGT results in a regulatory context for chemical status assessment is not considered as yet. However, passive sampling techniques are mentioned in Directive 2013/39/EU as “novel monitoring methods showing promise for future application”, and in three Technical Guidance Documents (Surface water chemical monitoring (10), Chemical monitoring of sediment and biota (11), and Biota monitoring (12)) as methods under development and evaluation that are desirable to introduce as they become available for improving the quality of the assessment. This opens the opportunity for their introduction in future directives if sufficient scientific evidence demonstrates their reliability for establishing the chemical status of water bodies.

Within the MONITOOL project, after exploring the relationships between dissolved and labile metal concentrations (13,14), a methodology has been proposed for the adaptation of EQS marine water to DGTs, for cadmium, nickel and lead by establishing linear model (15) and previous WP6 deliverable (16). This was accomplished by using a robust database of total dissolved and DGT-labile metal concentrations obtained by

concurrent sampling (i.e., spot sampling and DGT) in transitional and coastal waters of the eight European countries participating in the MONITOOL project (EAPA 565/2016) (Rodriguez et al., in prep.).

As part of the MONITOOL extension, an additional sampling campaign was done (2022) at sites known or suspected to be highly contaminated (in coastal area, estuary and closed basin) in order to complete the dataset in high concentrations with data close to or even above the EQS_{marine water} values. The objective was to increase the range of validity of the original models (equation 1) to include higher concentrations levels (near/above the EQS_{marine water}). In case the 2022 data doesn't improve the model, they can be used as "additional data" to check the fitness of the original MONITOOL models.

Scope

The scope of this deliverable is to update the MONITOOL data processing using the results of the 2022 campaign (additional campaign - Monitool extension).

This deliverable is a complementary work to the previous one (Amouroux et al., 2021 & 2023) (15,16), and it is advisable to review the previous one if better understanding of the methodological details is desired.

Field of application:

In this document, EQS refers only to AA-EQS and it does not apply to MAC-EQS. This document focuses on three metals and its compounds, defined as priority substances in the WFD: Ni, Pb, and Cd.

2. Methodology for using DGT results towards EQS_{marine water}

There are three options to use DGT labile concentration results in relation to EQS:

① Compare DGT results to an EQS_{bioavailable}

This option would be the best, as the labile fraction concentrate by DGTs appears to be the one that best predicts toxicity in biota, and thus appears very close to the (bio)available fraction (to the first link of the food chain). However, as no EQS_{bioavailable} is defined for Cd, Pb or Ni in marine waters, this option cannot be applied for the moment.

② Compare DGT results to an EQS_{DGT}

This option implies the adaptation of an EQS_{DGT} from the EQS_{marine water}.

Directive 2013/39/EU (3) states that an alternative biota taxon, or matrix, may be monitored instead of the specified matrix, **as long as the EQS applied provides at least an equivalent protection level.**

This implies to establish for each metal a valid relationship between the dissolved concentration measured in spot water samples and the DGT-labile concentration measured by DGTs (Figure 1). A relationship can be defined according to the following generic equation (equation 1):

$$[M]_{DGT} = \text{slope} \times [M]_{\text{Dissolved fraction}} + \text{intercept} \quad (\text{eq.1})$$

From equation 1, the EQS_{DGT} corresponding to the EQS_{marine water} value is determined, based on metal concentrations measured by both techniques and using a predictive model.

DGT results can then be compared to this EQS_{DGT}.

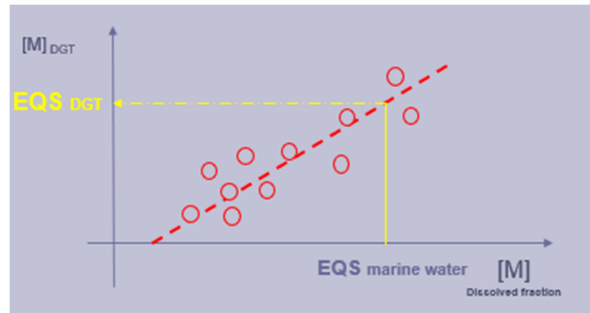


Figure 1: Illustration of the relationship $[M]$ in DGT labile and dissolved concentrations, and EQS_{DGT} adaptation
 $[M]_{DGT} = \text{slope} \times [M]_{\text{Dissolved fraction}} + \text{intercept}$

3 Compare DGT results to the EQS_{marine water}

This option implies to establish a valid relationship between the DGT labile and the dissolved concentrations measured in spot water samples, that allows the back-calculation of the dissolved concentrations from the measured DGT-labile concentrations. The relation is the inverse of the previous one (2nd possibility) and the relation can be described by equation 2: $[M]_{\text{Dissolved fraction}} = \text{slope} \times [M]_{DGT} + \text{intercept}$ (eq.2)

From equation 2, for each DGT result, a corresponding value is given in the dissolved fraction (Figure 2). This value can then be compared to the EQS_{marine water}.

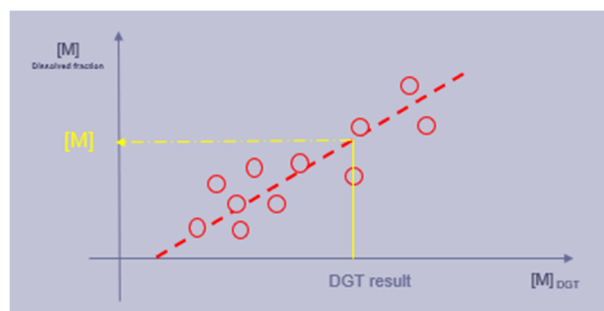


Figure 2: Illustration of the relationship $[M]$ in the dissolved fraction and in DGT,
 $[M]_{\text{Dissolved fraction}} = \text{slope} \times [M]_{DGT} + \text{intercept}$

Whether it is the 2nd or 3rd option, it is necessary to have results, acquired in parallel, from spot water samples and DGTs. The data must be reliable, homogeneous, and with low analytical uncertainty at the required low concentration levels. The sites sampled should cover a concentration range as wide as possible (from slightly contaminated to highly contaminated sites) and, if possible, present concentrations close or higher than the EQS value. For this purpose, sites suspected to be highly contaminated with Pb, Cd or Ni were targeted as a priority. This is why the MONITOOL partners have included port areas in their sampling sites. The data must be sufficiently numerous to allow obtaining a robust relationship.

In 2018, sampling campaigns were carried out by MONITOOL partners (AZTI, CEFAS, DCU, IFREMER, IPMA, ITC, MSS, SEPA and UNICA) covering eight countries (England, France, Ireland, Italy, Northern Ireland, Portugal, Scotland and Spain) located in five European sub-regions. Sampling campaigns were carried out, consisting of the simultaneous deployment of DGT samplers, collection of discrete water samples and measurement of physico-chemical parameters. A total of 36 sites were sampled in 2018 and when possible, the same stations were sampled in different seasons.

In 2022, within MONITOOL extension, an additional sampling campaign, was carried out by AZTI, CEFAS, DCU, Ifremer and Unica in 10 sites located in suspected highly contaminated sites, in estuary, in coastal area and in closed basin. The aim was to obtain results close to / above the EQS_{marine water} values in order to complete the 2018 data set so that the range of validity of the 2018 linear model would be wider.

The MONITOOL protocol is described in WP5 (17).

The following data processing is based on the MONITOOL dataset provided by WP4 (v24) updated with 2022 data. This dataset provides valid data results, after a validation process carried out by the expert laboratories in charge of the analysis.

Data processing:

For each metal, a statistical process has been run in order to identify outliers based on the results of linear models. The objective here is to identify as outliers those samples presenting standardized residuals, from the linear model, greater than 3 (rejection of values above 99.73% of the total values assuming that their distribution follows a normal distribution).

In this way, an iterative process is carried out until no more outliers are identified: the process starts with the linear modeling of all the results, thus allowing the identification of possible outliers. A new linear model is made by removing the previously identified outliers. The new outliers are then removed and the process is repeated until all the outliers are identified. The last model without outliers is then kept and diagnosed.

The diagnosis of the model is based on a standard graphical panel with graphical and numerical outputs to judge the normality and homoscedasticity of the residuals. The description of the outputs is done in Figure 8 for illustration purpose. Although there is a consensus on these parameters for the validation of linear models, the assumption of normality of the residuals is often considered secondary; the linear model is then considered to be robust to the absence of normality of the residuals given. Nevertheless, in addition to the QQ-plot (which allows a graphical analysis of the normality of the residuals) the Shapiro-Wilk and Lilliefors (Kolmogorov-Smirnov) tests were performed to guide the validation of the model (18). The most important thing is to have a QQ-plot curve that is as linear as possible with no real residuals that stand out (no curved distributed residuals). In this case, even if the results of the normality tests mentioned above are below the validity threshold (p -value < 0.05), the normality hypothesis is accepted (except when the p -value of the tests is far from the threshold).

The homoscedasticity of the residuals was also considered thanks to the graphical output illustrating the square roots of the residuals (standardized residuals) as a function of the predicted theoretical values (fitted-values) of Y and the Breush-Pagan and Golfeld-Quandt tests. Although we have taken care to use Koenker's version (less sensitive to the lack of normality than original version), the Breush-Pagan test stay sensitive to the lack of normality. The homoscedasticity of the residuals was also tested with the Golfeld-Quandt test.

The validity of the final model for each metal was evaluated, first based on its pValue (valid if ≤ 0.05), and secondly on the normality and homoscedasticity of the residues (valid if ≥ 0.05); It should nevertheless be specified that the choice to consider two tests for each criterion offers the possibility to be more objective on the decision (each test having its limits/gaps). Thus, the validation of a model is acquired if one of the 2 tests for each criterion is valid. As mentioned above, normality can be the subject of a less clear-cut decision, also based on the graphical output (QQ-plot). In any case, it is advisable to remain vigilant on this cut-off point of statistical thresholds (i.e. $p < 0.05$), keeping in mind the number of values taken into account, their general appearance by a visual glance (graphical output) and by relativizing the concept of "significant" (19).

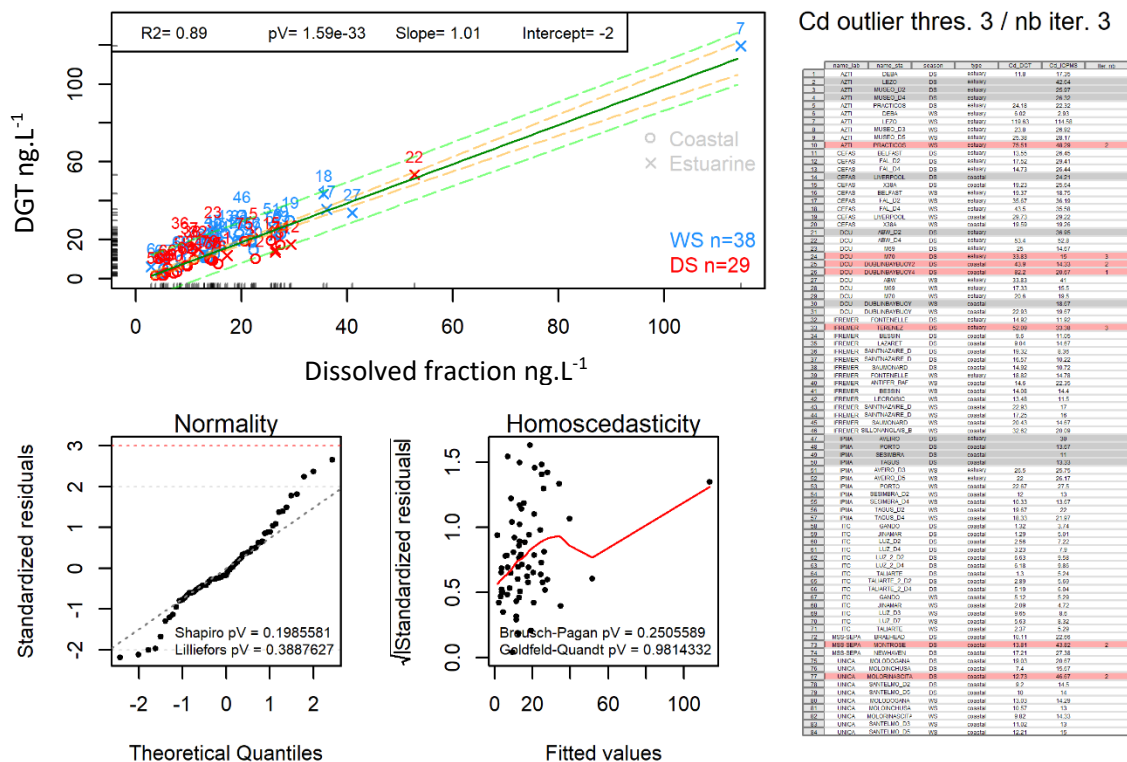


Figure 3: Illustration of standard output:

- Upper left chart show scatterplot $DGT=f(ICPMS)^*$ with linear model based on white results illustrated in the table on the right; the dark green line corresponds to the linear model, which is surrounded by its confidence interval CI 95 (orange dashed lines), and its predictive interval (PI 95) (red dashed lines).
*or $ICPMS=f(DGT)$;
- Bottom left chart represent QQ-plot illustrating normality of residuals with Shapiro and Lilliefors tests (normality: $pV > 0.05$);
- Bottom right chart represents Residuals variance plot illustrating homogeneity of residuals with Breusch-Pagan and Goldfeld-Quandt tests (homogeneity: $pV > 0.05$);
- Table on the right specifies the identity of samples with DGT and ICPMS values; background gray illustrate samples not considered because of one of DGT or ICPMS missing; background red color identify samples which are outliers; these two types of samples are not take into account in the regression. In some case, the number of iteration for outliers selection are specified.

Based on these models the EQS_{DGT} can then be determined. Even if the model is valid, its use is reliable within its validity range. Ideally this EQS_{DGT} is determined in the validity range. If this is not the case, it could be determined as the projection of the $EQS_{marine\ water}$ value ($EQS_{DGT} n^{\circ}1$), and to ensure that level of protection of

the EQS_{DGT} is the same or more than the EQS_{marine water}, it can be determined using the projection minus PI 95 (Prediction Interval 95%)(EQS_{DGT} n°2).

3. Determination of EQS_{DGT}

3.1. MONITOOL original model

MONITOOL data were used for establishing robust relationships between DGT labile and dissolved concentrations, allowing the determination of (A)-EQS_{DGT} for Cd, Ni and Pb. The graphical representations of the linear regression models between dissolved and labile concentrations for Cd, Ni and Pb (Equation 1) are presented in Figure 4 (15). Description of the equation 1 and the range of validity of the model are precised in Table 1.

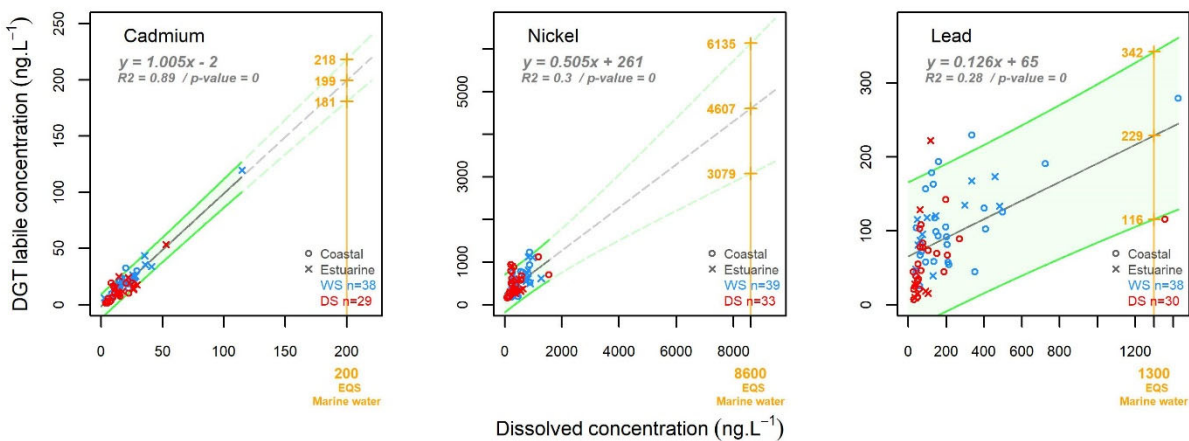


Figure 4: Linear regression models between dissolved and DGT labile concentrations for the determination of (A)-EQS_{DGT}: a) cadmium, b) nickel and c) lead. The regression line (black line) and its corresponding prediction interval ($\pm 95\%$; green lines and shaded green area) are shown. The shaded area corresponds to the range of validity of the model, defined on the horizontal axis, indicating that for a measured dissolved concentration value within the range of validity of the model, 95% of the DGT results will be between these 2 lines. WS: wet season, DS: dry season.

Table 1: Application of Equation 1 for predicting Cd, Ni and Pb DGT labile concentrations from dissolved concentrations. The mean and lowest predicted concentrations and the validity range are shown.

	Mean predicted $[M]_{DGT}$ (ng·L ⁻¹)	Lowest predicted $[M]_{DGT}$ (ng·L ⁻¹) (LPI 95%)	Validity range (ng·L ⁻¹)
Cadmium	$[Cd]_{DGT} = 1.01 [Cd]_{Dissolved\ concentration} - 2$	$[Cd]_{DGT} = 0.98 [Cd]_{Dissolved\ concentration} - 12$	$[Cd]_{Dissolved\ concentration}$ [<LQ; 114]
Nickel	$[Ni]_{DGT} = 0.51 [Ni]_{Dissolved\ concentration} + 261$	$[Ni]_{DGT} = 0.48 [Ni]_{Dissolved\ concentration} - 166$	$[Ni]_{Dissolved\ concentration}$ [<LQ; 1,544]
Lead	$[Pb]_{DGT} = 0.13 [Pb]_{Dissolved\ concentration} + 65$	$[Pb]_{DGT} = 0.11 [Pb]_{Dissolved\ concentration} - 31$	$[Pb]_{Dissolved\ concentration}$ [<LQ; 1,428]

3.2. Update of the models considering additional campaign 2022 data

The dataprocessing was carried out using 2018 and 2022 results (WP5 valid data – WP4). A comparison of the 2022 model (including both 2018 and 2022 data) to the original model is done Figure 5. Table 2 provides details of the changes in DGT_{EQS} n°1 and 2, the range of validity and the R2 of the model using 2022 data compared with the original model.

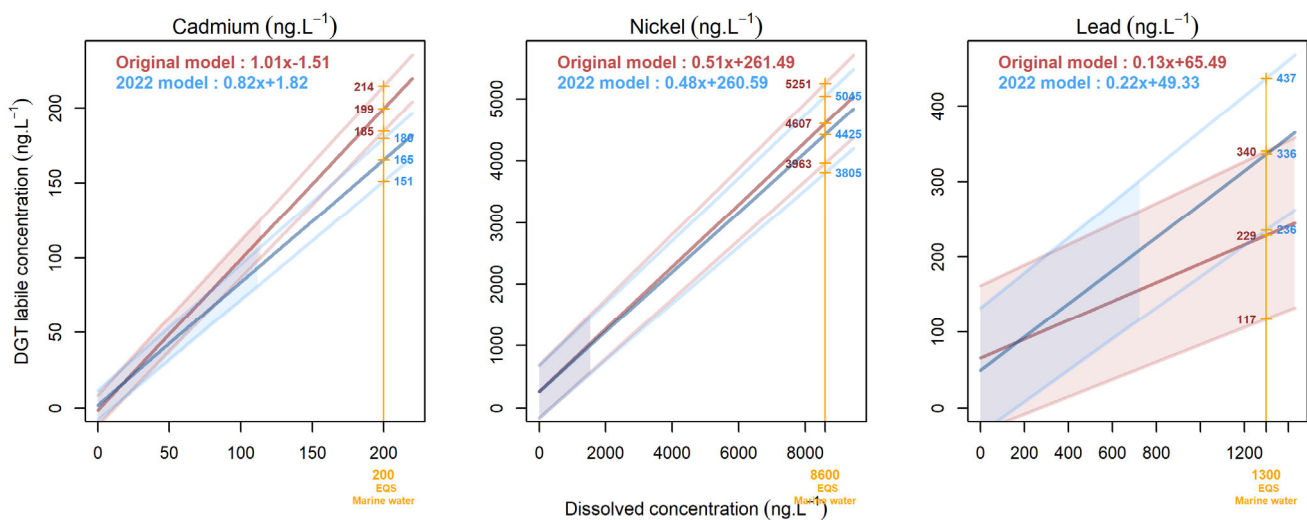


Figure 5: Comparison of the linear regression models between dissolved and DGT labile concentrations for the determination of (A)-EQS_{DGT} using 2018 and 2022 data (in blue line) to the original models (red line) a) cadmium, b) nickel and c) lead. The regression line (dark line) and its corresponding prediction interval (±95%; light lines and shaded area) are shown. The shaded area corresponds to the range of validity of the model, defined on the horizontal axis, indicating that for a measured dissolved concentration value within the range of validity of the model, 95% of the DGT results will be between these 2 lines.

Table 2: Comparison of EQS_{DGT} adaptation, range of validity and R2 of the updated model (2018 and 2022 data) to the original model (2018 data)

Metal	EQS _{DGT} n°1 µg.L ⁻¹	EQS _{DGT} n°2 µg.L ⁻¹	EQS _{DGT} n°1 bis/ « new » µg.L ⁻¹	EQS _{DGT} n°2 bis/ « new » µg.L ⁻¹	Extension of the validity range	Improvement of the R2
Cadmium	0.20	0.18	0.16	0.15	no	R2: 0.89 → 0.85
Nickel	4.60	3.08	4.42	3.81	no	R2: 0.3 → 0.29
Lead	0.23	0.12	0.34	0.24	no	R2: 0.28 → 0.33

The additional campaign (2022 data) does not allow to improve the model, either the range of validity or the R2, so the option of using 2022 data to refine the original model is not retained.

It should be underlined here that the data processing identifies “high” concentrations as outliers, since there is very few data in “high” concentrations levels (near or above the EQS_{marine water} values for cadmium and Nickel). It will therefore be very difficult to increase the validity range of the model as the concentrations measured in the marine environment are very similar to those measured in MONITOOL.

3.3. Use additional campaign 2022 data to verify original models

To check the reliability and relevance of the original models, additional 2022 data (DGT - ICPMS) were integrated (purple cross); graphs with those additional data are presented Figure 6.

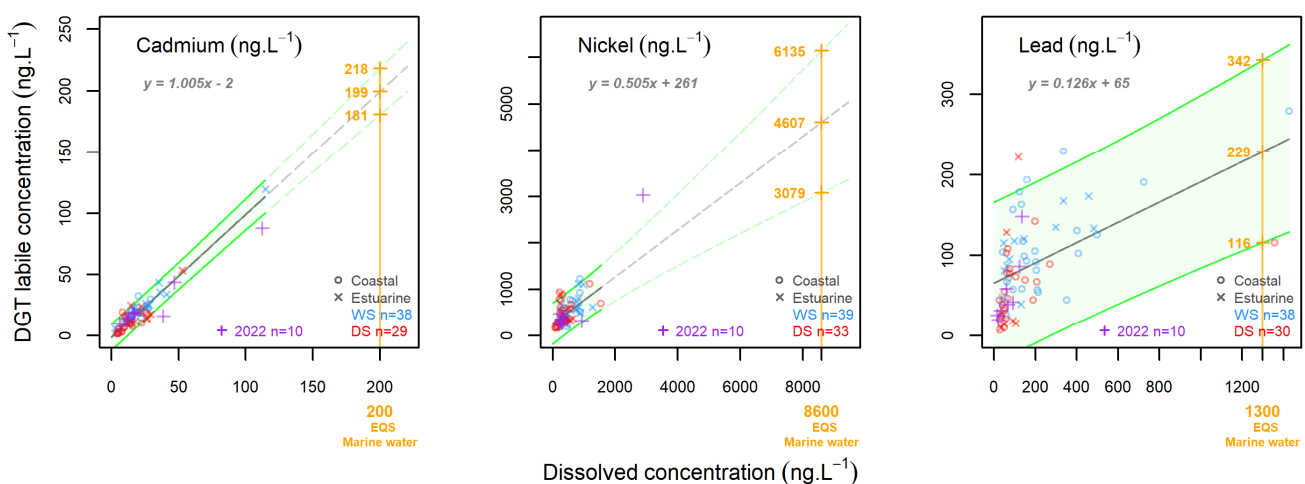


Figure 6: Superposition of the 2022 data (purple cross) to the original MONITOOL linear regression models between dissolved and DGT labile concentrations a) cadmium, b) nickel and c) lead. The regression line (black line) and its corresponding prediction interval (±95%; light green lines and shaded area) are shown. The shaded area corresponds to the range of validity of the model, defined on the horizontal axis, indicating that for a

measured dissolved concentration value within the range of validity of the model, 95% of the DGT results will be between these 2 lines.

Considering data from the additional 2022 MONITOOL campaign, all the data fitted the regression line except 2 results for Cd (results are just below the lower regression line (-PI 95%)) and 1 result for Ni (above the higher lower regression line (+PI 95%)).

5. Main conclusion

As previously mentioned, this study should be considered as complementary to Amouroux et al. 2021 and 2023 (15,16). The additional MONITOOL sampling campaign 2022 was done in order to get results in high concentrations (near or above the EQS_{marine water} values) to extend ideally the range of validity of the original models for cadmium, nickel and lead. The data obtained, although targeting sites suspected of being highly contaminated, did not make it possible to reach such levels, with the exception of Bassin sites (confined Basin), which is not representative of concentrations that can be found in the marine environment. The modelling process excludes data considered to be outliers, as incorporating them would have too great influence on the model. It is therefore clear that unless a large amount of data is acquired at high levels (close to the EQS_{marine water} value), it will not be possible to extend the model's range of validity using field data. However, the use of 2022 data enable us to check that the original MONITOOL models do indeed fit.

Reference

1. E.C. Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy. Official Journal of the European Union L327/1, 22.12.2000. 2000;
2. E.C. Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European Parliament and of the Council. Official Journal of the European Union. 24.12.2008. L 348/84. 2008;
3. U.E. Directive 2013/39/EU of the European Parliament and of the council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy. Official journal of the European Union. 24.8.2013. L 226/1. 2013;
4. OSPAR Commission. CEMP Guidelines for Monitoring Contaminants in Biota (Agreement 1999-02). Revision 2018. OSPAR Commission [Internet]. :126. Available from: <https://www.ospar.org/documents?d=32414>
5. OSPAR Commission. CEMP Guidelines for Monitoring Contaminants in Sediments (Agreement 2002-15). Revised in 2018/2019. OSPAR Commission [Internet]. :118. Available from: <http://www.ospar.org/documents?d=32743>
6. Menegário AA, Yabuki LNM, Luko KS, Williams PN, Blackburn DM. Use of diffusive gradient in thin films for in situ measurements: A review on the progress in chemical fractionation, speciation and bioavailability of metals in waters. *Anal Chim Acta*. 2017 Aug 29;983:54–66.
7. Davison W, Zhang H. In situ speciation measurements of trace components in natural waters using thin-film gels. *Nature* [Internet]. 1994 Feb;367(6463):546. Available from: <https://www.nature.com/articles/367546a0>
8. Zhang Hao, Davison William. Performance Characteristics of Diffusion Gradients in Thin Films for the in Situ Measurement of Trace Metals in Aqueous Solution. *Anal Chem* [Internet]. 1995 Oct 1;67(19):3391–400. Available from: <https://doi.org/10.1021/ac00115a005>
9. Zhang H, Davison W. In situ speciation measurements. Using diffusive gradients in thin films (DGT) to determine inorganically and organically complexed metals. *Pure Appl Chem - PURE APPL CHEM*. 2001 Jan 1;73:9–15.
10. E.C. CIS Guidance Document n°19. Guidance on surface water chemical monitoring under the water framework directive. Technical Report 2009 - 025. 2009;132.
11. European Commission. Directorate General for the Environment. Guidance on chemical monitoring of sediment and biota under the water framework directive. Guidance document No 25 . [Internet]. LU: Publications Office; 2010 [cited 2021 Jun 25]. Available from: <https://data.europa.eu/doi/10.2779/43586>
12. E.C. CIS Guidance Document n°32 on biota monitoring (the implementation of EQSbiota) under the water framework directive. Technical Report 2014 - 083. 2014;87.
13. Rodríguez JG, Amouroux I, Belzunce-Segarra MJ, Bersuder P, Bolam T, Caetano M, et al. Assessing variability in the ratio of metal concentrations measured by DGT-type passive samplers and spot sampling in European seawaters. *Sci Total Environ* [Internet]. 2021 Aug [cited 2021 Nov 8];783:147001. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0048969721020714>
14. Caetano M, Correia dos Santos MM, Rosa N, Carvalho I, Rodríguez JG, Belzunce-Segarra MJ, et al. Metals concentrations in transitional and coastal waters by ICPMS and voltammetry analysis of spot samples and passive samplers (DGT). *Mar Pollut Bull* [Internet]. 2022 Jun [cited 2022 Jun 29];179:113715. Available from: <https://linkinghub.elsevier.com/retrieve/pii/S0025326X22003976>

15. Amouroux I, Gonzalez JL, Guesdon S, Belzunce-Segarra MJ, Bersuder P, Bolam T, et al. A new approach to using Diffusive Gradient in Thin-films (DGT) labile concentration for Water Framework Directive chemical status assessment: adaptation of Environmental Quality Standard to DGT for cadmium, nickel and lead. *Environ Sci Eur* [Internet]. 2023 Apr 29 [cited 2023 Jul 11];35(1):29. Available from: <https://enveurope.springeropen.com/articles/10.1186/s12302-023-00733-4>
16. Amouroux I, Gonzalez JL, Guesdon S, Menet-Nédélec F, Dallet M. DGT : EQS adaptation for Nickel, Cadmium and Lead . MONITOOL project. Technical deliverables. WP6. Action 1. 2021.
17. Bolam T, Bersuder P. Monitool- Sampling protocol. Cefas. 20p. 2018.
18. Knief U, Forstmeier W. Violating the normality assumption may be the lesser of two evils. *bioRxiv* [Internet]. 2020 May 5 [cited 2020 Sep 28];498931. Available from: <https://www.biorxiv.org/content/10.1101/498931v2>
19. Wasserstein RL, Schirm AL, Lazar NA. Moving to a World Beyond “ $p < 0.05$ ”. *Am Stat* [Internet]. 2019 Mar 29 [cited 2020 Sep 28];73(sup1):1–19. Available from: <https://www.tandfonline.com/doi/full/10.1080/00031305.2019.1583913>

