Biocidal Products Committee (BPC)

Opinion on a request according to Article 75(1)(g) on

The evaluation of dioxins emissions from the use of the biocidal product family (BPF) "CMIT/MIT SOLVENT BASED" in fuels used in road and ship transport

ECHA/BPC/283/2021

Adopted

17 June 2021

P.O. Box 400, FI-00121 Helsinki, Finland | Tel. +358 9 686180 | echa.europa.eu

Opinion of the Biocidal Products Committee

on the evaluation of dioxins emissions from the use of the biocidal product family (BPF) "CMIT/MIT SOLVENT BASED" in fuels used in road and ship transport

In accordance with Article 75(1)(g) of Regulation (EU) No 528/2012 of the European Parliament and of the Council 22 May 2012 concerning the making available on the market and use of biocidal products, the Biocidal Products Committee (BPC) has adopted this opinion on the evaluation of dioxins emissions from the use of the biocidal product family (BPF) "CMIT/MIT SOLVENT BASED" in fuels used in road and ship transport.

This document presents the opinion adopted by the BPC, having regard to the conclusions of the rapporteur.

Process for the adoption of the opinion

A request by Commission was received by ECHA on 24 of July 2020. The request was confirmed by ECHA to be passed to the BPC by 27 July 2020. The BPC appointed the rapporteur at its meeting of 6-8 October 2020 (BPC-36). The rapporteur presented the draft opinion to the BPC at its meeting of June 2021.

A draft report was provided to the BPC members and the applicant for consultation on 26 March 2021, with a deadline for providing comments of 14 April 2021. Comments provided during this consultation were incorporated. The report forms the basis for the opinion and is provided as an Annex.

Following the adoption of the opinion at the BPC meeting of 15-18 June 2021 the opinion was amended accordingly and delivered by ECHA to the Commission on 6 July 2021.

Adoption of the opinion

Rapporteur: France

The BPC opinion on the evaluation of dioxins emissions from the use of the biocidal product family (BPF) “CMIT/MIT SOLVENT BASED” in fuels used in road and ship transport was adopted on 17 June 2021.

The BPC opinion was adopted by consensus*.* The opinion is published on the ECHA webpage at: https://echa.europa.eu/regulations/biocidal-products-regulation/approval-of-active-substances/opinions-on-article-75-1-g

Further details of the opinion and background

1. Request for the opinion and background

During the 34th Biocidal Products Committee (BPC) meeting of March 2020, ECHA adopted its final opinion on the application for union authorisation of the biocidal product family "C(M)IT/MIT SOLVENT BASED" for use as preservative in aviation fuel, crude oil and middle distillate fuel (product type 6).

During this 34th BPC meeting, the consequence of the presence of halogenated organic compounds, such as C(M)IT/MIT in fuel for automotive use, on the formation of dioxins was questioned. Due to a nation law prohibiting in their territory the addition of chlorine or bromine compounds as fuel additives which could lead to formation of dioxins via combustion, the member from the German Competent Authority expressed a minority opinion and announced that it wants to request the Commission to decide in accordance with Article 44(5) of the BPR that a Union authorisation for this biocidal product family shall not apply in their territory. Therefore, the Commission considered that it should be clarified whether the products in the family lead to the generation of dioxins to an extent that they would have unacceptable effects on human health or the environment, and whether authorising this biocidal product family would be in line with the objectives set in Regulation (EU) 2019/1021. Therefore, the Commission mandated ECHA under Article 75(1)(g) of the BPR to provide an opinion on the "Estimation of the emissions of dioxins from the use of the "C(M)IT/MIT SOLVENT BASED" BPF as preservative in crude oil and middle distillate fuel".

Dioxins and dioxin-like substances, including Polychlorinated Biphenyls (PCBs), are persistent organic pollutants (POPs) covered by the Stockholm Convention[1] and the Aarhus Protocol[2]. Regulation (EU) 2019/1021 (the POP Regulation)[3] was adopted to implement the Union's obligation under the previously mentioned protocol and convention. The objective of Regulation (EU) 2019/1021 is to protect human health and the environment from POPs by prohibiting, phasing out as soon as possible, or restricting the manufacturing, placing on the market and use of substances subject to the Stockholm Convention or the Aarhus Protocol. The Commission considered in the mandate that it should be clarified whether, due to the formation of dioxins from C(M)IT/MIT during fuel combustion, the biocidal product family (BPF) "C(M)IT/MIT SOLVENT BASED" has no unacceptable effects on human health and environment.

First, two initial issues were raised around the need to:

a. estimate the amount of formation of dioxins (in mg/year TEQ (toxic equivalents)) due to the use of "C(M)IT/MIT SOLVENT BASED" in fuels used for road and water transport at the maximum application rate proposed for authorisation and the highest amount of product produced per year according to the authorisation. [redacted]

b. estimate the overall contribution to the emissions of dioxins coming from the use of "C(M)IT/MIT SOLVENT BASED" as preservative for fuels used in road and water transport in the worst –case scenario, and compare it, where data are available, to the emissions of dioxin coming from other sectors or sources.

[1] http://www.pops.int/
[2] https://www.unece.org/env/lrtap/pops_h1.html
[3] https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019R1021&from=EN

Once these estimations have been made, two additional issues raised by the Commission need to be handled:

c. Clarify the level of the risks for human health due to the exposure via the environment, and more specifically through air pollution, by:

1. Assessing the contribution that the use of "C(M)IT/MIT SOLVENT BASED" will have in the exposure to dioxins.
2. Assessing the level of risks for human health, either by a quantitative assessment or a qualitative assessment.
3. Providing an opinion on whether the risks can be considered acceptable or not, with or without setting specific/conditions to the authorisation.

d. Clarify the level of the risks to the environment by:

1. Assessing the level of risks for the environment, either by a quantitative assessment or a qualitative assessment, considering the estimation of the contribution to the emission of dioxins from the use of "C(M)IT/MIT SOLVENT-BASED" as preservative for fuels used in road and water transport.
2. Providing its opinion on whether the risks can be considered acceptable or not, with or without setting specific/conditions to the authorization.

2. Summary of information supporting the request for the opinion

2.1. Aim and content of the report

To answer these questions, a bibliographic review on the formation of dioxins (PCDD/F[4]) due to road traffic[5] and theoretical calculations on the potential formation of dioxins that could result from the combustion of C(M)IT/MIT were undertaken.

An analysis of the scientific literature focused on C(M)IT/MIT was first performed then enlarged to tackle two broader questions:

1. What is the amount of dioxins (PCDD/F) formed by road traffic, in particular due to addition of halogenated compounds in fuels,

2. What is the contribution of dioxins due to road traffic to total dioxin emission from all sources?

This extension of the scope of the bibliographic review aimed at assessing if the dioxin emission from road traffic/chlorine compounds could be considered as negligible and, consequently, the initial concern as minor.

[4] The generic term "dioxins" refers here –in the whole text- to Polychlorinated dibenzo(p)dioxins and furans. (PCDD/F).

[5] The Commission mandate requested an estimation of the amount of formation of dioxins and the overall contribution to the emissions of dioxins due to the use of "C(M)IT/MIT SOLVENT BASED" in fuels used for road and water transport. However, due to the lack of public scientific data relating to water transport, the report focuses only on road traffic.

2.2. Methodology

The review of the scientific literature was conducted using Scopus and PubMed library database, according to the main principles of the EFSA guidance on systematic review methodology (EFSA, 2010). It was completed by additional searches following snowball approach and a search on Google Scholar with keywords similar to those considered for Scopus and PubMed search.

All the elements mentioned in the chapter 3 of the Mandate of the EU Commission according to Art.75 of the BPR (ARES (2020)3922032 – 24/07/2020), to be considered for addressing this question, were also included in the analysis, as well as reports mentioned along the exchange with the other Member States.

2.3. Outcome of the bibliographic synthesis

2.3.1. The formation of dioxins from the use C(M)IT/MIT in fuels

No publication concerning the use of C(M)IT/MIT in fuels and the formation of dioxins was retrieved from the search. It was therefore not possible to answer any of the questions raised by the EU Commission (i) on the amount of formation of dioxins from the use C(M)IT/MIT in fuels and its contribution to total emissions of dioxins, and consequently, (ii) on the risks associated with this use for human health and for the environment.

2.3.2. The formation of dioxins from road traffic and the impact of adding chlorine compounds in fuel on dioxin emission

The formation of dioxins through combustion processes has been thoroughly described in the scientific peer reviewed literature. It occurs when four important process conditions are met: (1) temperature between 200-800 °C (ideal range between 200-400 °C), (2) presence of chlorine, (3) presence of organic (specifically aromatic) matter and (4) presence of oxygen.

Besides, in some specific cases (exhaust after-treatments or fuel additives), the presence of metals (which serve as catalysts, e.g. copper) could also affect strongly the formation of dioxins.

Overall, there is no doubt from literature that motor vehicles powered with fuels containing chlorine are sources of dioxin and furan emission but the magnitude of their total input remains uncertain.

2.3.3. Estimation of PCDD/F emission variability

Literature review reveals that emission factors show very large variability (by over a factor of 200), even for the same car under comparable test conditions.

This variability can be attributed to many factors: measurement methods and approach (laboratory/tunnel/on road; particulate/gaseous phase; detection limits), operating conditions (engine speed rate cycle), compounds identity and toxic equivalence pattern, fuel and oil characteristics (especially scavenger content), engine characteristics.

Due to methodological differences, all these values are not strictly comparable but give the range of the main differences between fuel types (leaded petrol, unleaded petrol, diesel) and vehicle technology types (e.g. non-catalyst passenger cars, catalyst passenger cars, light-duty vehicles, heavy-duty vehicles). It is worth to note that they are representative of various cases throughout the world, from eighties to 2010 but would need to be updated to describe more precisely the potential emission from current fleet and fuel uses in Europe.

Overall data show higher emissions for leaded petrol, which are mainly attributed to the “scavenger-compounds” added to the fuels to prevent the engine from the formation of lead precipitates. The level of emission from diesel is less clear and its potential contribution to total dioxin emissions is still controversial.

2.3.4. Impact of adding chlorine compounds in fuel on dioxin emission

Chlorine is a key precursor of the dioxin formation from combustion. Although well studied, the link between fuel chlorine content and the level of dioxin formation is however far from clearly described. Strong correlations between chlorinated additives in petrol or engine oil and PCDD/PCDF emissions were shown by some studies. Similarly, reduction of PCDD/PCDF and PCB emissions was reported when halogens, especially chlorine, are excluded from fuels and oils. This relationship was presented as the main explanation of the high dioxin emission from leaded petrol, which were characterized by higher chlorine content due to the addition of halogenated additives or "scavengers". The use of chlorinated additives in fuels was thus suspected to be a key factor driving dioxin formation in vehicle engines, leading some European countries to regulate their content in fuels.

The linear relationships between chlorine content and dioxin formation has however not been observed as a rule for various fuels and vehicle technology types.

Several experiments have shown that chlorine content has no or nonlinear impact on the synthesis of PCDD/Fs, while others have shown that fuel doped to high levels of chlorine, especially when catalytic conditions occur, can significantly increase the formation of PCDD/Fs for diesel fuels. Reliable description of the relationship between fuel chlorine content and dioxin emission from fuel combustion (i.e. mathematical model with validity domains) cannot be derived from scientific literature.

Hence, estimation of dioxin emissions cannot be merely derived from maximum level of chlorine contribution to the fuel from C(M)IT/MIT and conversion yield of chlorine calculated from publications.

2.3.5. Contribution of traffic to total dioxin emission from all sources

Emissions from road traffic were estimated to be in the range of 0.2 % to 12.3 % with most of the estimations under 3 % and a high variability from one country to another.

This discrepancy is not only due to differences of the specificities of countries in terms of road traffic or industrial equipment but also to the differences between methods followed to obtain these estimations and to the uncertainty surrounding each individual estimation of dioxin emission. All these estimations are characterized by very large uncertainties and much of the data used to estimate road traffic emissions in these inventories date back 15-20 years. Their relevance to describe current European situation shall be investigated.

PCDD/F emissions from road traffic have experienced a significant decrease of dioxin emissions from road transport since 1995 due to the reduction of the lead content in petrol and the decrease of the use of scavengers. In the meantime, higher reductions were observed for the dominant industrial sources (e.g. municipal solid waste or industrial incinerators), due to the implementation of new regulation and technical progress. Nowadays, although still minor compared to current dominant sources, the relative proportion of non-industrial diffuse sources, including transport, in total emissions is consequently likely to increase.

Besides the uncertainty surrounding these estimations and the rapid evolution of automobile industry (new fuels, engines and after-treatment technologies) prevent, for now, to draw a relevant state of the situation and simple meaningful conclusions. New after-treatment technology and biofuels or doped fuels cannot be invoked, for the moment, as the absolute solution to limit dioxin emission from traffic.

Although emissions from traffic are minor compared to current dominant sources, it should be noted that their relative importance in terms of relative human exposure could be higher, due to their spatial distribution in densely populated urban areas, close to the human population and living environment and with less dilution of emissions.

This review has been focused on dioxin emissions from road traffic whatever their origin. It should be noted however that the use of C(M)IT/MIT in fuel is not restricted to automotive fuels but can includes also fuels for residential heating. Such use shall be considered in the estimation of the potential amount of formation of dioxins resulting from the use of "C(M)IT/MIT SOLVENT BASED" in fuels.

2.4. Theoretical calculation on the potential formation of dioxins from the combustion of C(M)IT/MIT in fuels

One Member State has provided a theorical calculations in order to check if the use, as prescribed in the SPC of the biocidal product family "C(M)IT/MIT SOLVENT BASED", can lead to unacceptable concentrations of PCBs and dioxins in fuels, according to its national legislation.

It was concluded from these theoretical calculations that the use, as prescribed in the SPC of the product, may lead to concentrations of PCBs and dioxins in fuels above national legal thresholds.

This calculation does not allow to estimate the amount of dioxins formed due to the use of "C(M)IT/MIT SOLVENT BASED" in fuels.

Theoretical calculation of dioxin emissions derived from the use of the products of C(M)IT/MIT SOLVENT-BASED family for preservation of fuel has also been proposed by the applicant.

This theoretical calculation assumes conversion of moles of chlorine atoms, [redacted] into dioxin, based on linear relationship assumed between chlorine content and dioxin formation. The conversion yield of chlorine into TCDD equivalents (TEQ) is taken from a study published by Marklund *et al.* (1990)[6].

Total dioxin emission was then estimated by calculating the yearly total volume of fuel necessary for a treatment with the whole annual tonnage of C(M)IT/MIT solvent for Europe, at the dose considered for curative treatment (3-6 ppm of C(M)IT/MIT).

This approach results in a total maximum emission [redacted] mg/year TEQ in the whole of Europe from the use of the C(M)IT/MIT solvent as preservative in crude oil and middle distillate fuel. This estimation is deemed by the applicant as relevant for essentially all motor classes except diesel engines with copper-catalyzed particulate traps.

These calculations present several severe shortcomings. The main is the assumption of the linear relationship between chlorine content and dioxin formation, while the review of the scientific literature has shown that such relationship has not been observed as a rule for the various fuel and vehicle technology types. Besides the dioxin emissions collected from literature show that (i) very few couple of values for chlorine content / dioxin emission are available, (ii) numerous studies are representatives of outdated fuels/engines and partly

[6] Marklund, S., Andersson, R., Tysklind, M., Rappe, C., Egebäck, K.E., Björkman, E. and Grigoriadis, V. (1990). Emissions of PCDDs and PCDFs in gasoline and diesel fueled cars. Chemosphere 20: 553–561.

specific to non-EU countries. Consequently, a reasonable worst-case conversion yield representative of current EU fuels and vehicle technology types cannot be derived from the literature and the final estimation reported above is not agreed.

An attempt of worst-case theoretical estimation of the dioxin emissions resulting from the use claimed for the products C(M)IT/MIT solvent-based has at last been proposed. The calculation is based on four assumptions: (1) a total conversion of moles of chlorine atoms into dioxin or PCB, (2) each chlorine atom from the product allows the formation of the most toxic dioxin is 2,3,7,8-TCDD (2,3,7,8-tetrachlorodibenzo-p-dioxin), (3) chlorine atoms are provided by [redacted] and (4) the total dioxin emission is resulting from the use of the whole annual tonnage of C(M)IT/MIT solvent in Europe. Several worst-case assumptions were also considered for the selection of parameters (e.g. application rate).

The total of dioxin emission estimated by this approach is clearly overestimated and has no real meaning compared to available estimations of the total dioxin emission in Europe (Quass et al. 2000). The overestimation is mainly due to the assumption of a total conversion of moles of chlorine atoms into 2,3,7,8-TCDD. In the absence of reliable parameters (i.e. consistent with the scientific literature) to describe the relationship between fuel chlorine content and dioxin emission from fuel combustion, it is not possible to provide a realistic estimation of the dioxin emission.

3. Overall conclusions

The review of scientific literature shows the absence of peer-reviewed publications on the potential of dioxin formation resulting from the use of C(M)IT/MIT as preservative in oil and fuel. There is no experimental evidence demonstrating the existence or the absence of formation of dioxin from the use of C(M)IT/MIT as preservative in oil and fuel. This phenomenon is likely to occur, but the extent cannot be estimated.

Motor vehicles are significant sources of emission of dioxins, but minor compared to current dominant sources. However, along with the decrease of dominant industrial sources of dioxins, the relative proportion of non-industrial diffuse sources, including transport, in total emissions is likely to increase, at least in some EU countries. Overall, high uncertainties are surrounding estimations of dioxins emissions from traffic due to the actual scarcity of measurements with respect to the numerous key factors involved in the process (e.g. fuel and oil characteristics as detailed above), difference in the methodology used for estimations and analytical measurements. Besides, the global situation behind this issue is highly dynamic (vehicle fleet, traffic level), which leads to some data being partially outdated and therefore the final interpretation uncertain.

The addition of chlorine, through halogenated additives or "scavengers", in the fuel increases dioxin formation under certain conditions (e.g. incomplete combustion as observed in vehicle engines). However, relationship between chlorine content and dioxin formation is not yet clarified and does not seem to be broadly linear.

Hence, although the potential formation of dioxin from the use of C(M)IT/MIT as preservative in oil and fuel is likely to occur, it is not possible to draw any conclusions either on the magnitude of the potential contribution of the use of C(M)IT/MIT in fuels with respect to dioxin exposure, nor on the potential consequence of chlorine additive such as C(M)IT/MIT in fuels on human health and on environment. None of the initial questions raised by the European Commission can therefore be addressed.

In order to address the questions as asked by the Commission further experiments should be performed to provide factual data demonstrating that the use of C(M)IT/MIT SOLVENT could be a source of dioxins and to measure to what extent.

Annexes

Response to the EU Commission mandate requesting ECHA opinions under Article 75(1)(g) of the BPR

"Estimation of the emissions of dioxins from the use of the "CMIT-MIT SOLVENT BASED" biocidal product family (BPF) as preservative in crude oil and middle distillate fuel"

I. Context

During the 34th Biocidal Products Committee (BPC) meeting of March 2020, ECHA adopted its final opinion on the application for union authorisation of the biocidal product family "C(M)IT/MIT SOLVENT BASED" for use as preservative in aviation fuel, crude oil and middle distillate fuel (PT 6).

During this 34th BPC meeting, the consequence of the presence of halogenated organic compounds, such as C(M)IT/MIT in fuel for automotive use, on the formation of dioxins was questioned.

Dioxins and dioxin-like substances, including Polychlorinated Biphenyls (PCBs), are persistent organic pollutants (POPs) covered by the Stockholm Convention[1] and the Aarhus Protocol[2]. Regulation (EU) 2019/1021 (the POP Regulation)[3] was adopted to implement the Union's obligation under the previously mentioned protocol and convention. The objective of Regulation (EU) 2019/1021 is to protect human health and the environment from POPs by prohibiting, phasing out as soon as possible, or restricting the manufacturing, placing on the market and use of substances subject to the Stockholm Convention or the Aarhus Protocol. Therefore, the European Commission considers that it should be clarified whether, due to the formation of dioxins from C(M)IT/MIT during fuel combustion, the biocidal product family (BPF) "C(M)IT/MIT SOLVENT BASED" has no unacceptable effects on human health and environment.

In this framework, on July 2020, the EU Commission mandated ECHA under Article 75(1)(g) of the BPR (ARES (2020)3922032 – 24/07/2020) to provide an opinion on the "Estimation of the emissions of dioxins from the use of the "C(M)IT/MIT SOLVENT BASED" BPF as preservative in crude oil and middle distillate fuel". Two initial issues were raised around the need to:

a. estimate the amount of formation of dioxins (in mg/year TEQ (toxic equivalents)) due to the use of "C(M)IT/MIT SOLVENT BASED" in fuels used for road and water transport at the maximum application rate proposed for authorisation and the highest amount of product produced per year according to the authorisation. [redacted]
b. estimate the overall contribution to the emissions of dioxins coming from the use of "C(M)IT/MIT SOLVENT BASED" as preservative for fuels used in road and water transport in

[1] http://www.pops.int/
[2] https://www.unece.org/env/lrtap/pops_h1.html
[3] https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32019R1021&from=EN

the worst –case scenario, and compare it, where data are available, to the emissions of dioxin coming from other sectors or sources.

Once these estimations have been made, two additional issues raised by the Commission need to be handled:

c. Clarify the level of the risks for human health due to the exposure via the environment, and more specifically through air pollution, by:

1. Assessing the contribution that the use of “C(M)IT/MIT SOLVENT BASED” will have in the exposure to dioxins.

2. Assessing the level of risks for human health, either by a quantitative assessment or a qualitative assessment.

3. Providing an opinion on whether the risks can be considered acceptable or not, with or without setting specific/conditions to the authorisation.

d. Clarify the level of the risks to the environment by:

1. Assessing the level of risks for the environment, either by a quantitative assessment or a qualitative assessment, considering the estimation of the contribution to the emission of dioxins from the use of “C(M)IT/MIT SOLVENT-BASED” as preservative for fuels used in road and water transport.

2. Providing its opinion on whether the risks can be considered acceptable or not, with or without setting specific/conditions to the authorization.

Following the receipt of the Mandate of the EU Commission according to Art.75 of the BPR, ECHA requested on July 2020, France, as e-CA of Union Authorisation for the biocidal product family “C(M)IT/MIT SOLVENT BASED”, to prepare the report aiming to discuss and reply to the questions raised in the Mandate. This is the purpose of this document.

II. Aim and content of this report

To answer the questions from the EU Commission, this report contains two complementary sections: a bibliographic review on formation of dioxins (PCDD/F[4]) due to road traffic (Chapter IV) and theoretical calculations on the potential formation of dioxins that could result from the combustion of C(M)IT/MIT (Chapter V).

An analysis of the scientific literature focused on C(M)IT/MIT was first performed. Then the scope was enlarged to tackle two broader questions:

1. What is the amount of dioxins (PCDD/F) formed by road traffic, in particular due to addition of halogenated compounds in fuels,
2. What is the contribution of dioxins due to road traffic to total dioxin emission from all sources?

The aim of this reframing is not to draw up an extensive review. It is rather to shed light on the initial issues and see if the initial concern could be considered as minor if the literature review concludes that the dioxin emission from road traffic/chlorine compounds is negligible.

In chapter V, theoretical calculations provided by NL CA (Ctgb) and the applicant on the potential formation of dioxins that could result from the combustion of the chlorine input by C(M)IT/MIT SOLVENT

[4] In this report the generic term “dioxins” refers to Polychlorinated dibenzo(p)dioxins and furans. (PCDD/F).

in fuel are presented. Additional theoretical calculations considering all chlorine input brought by C(M)IT/MIT SOLVENT in fuel are also proposed following a worst-case approach.

III. Methodology

1. Data sources

Several data collection techniques have been used.

First, a review of the scientific literature was conducted using Scopus and PubMed library database, according to the main principles of the EFSA guidance on systematic review methodology (EFSA, 2010)[5] .

Then, additional searches were performed following snowball approach (search of additional articles cited as key publications in the set of articles retrieved by the initial search) completed by free search on Google Scholar using keywords similar to those considered for Scopus and PubMed search.

In addition to this, the following source of potentially relevant information was also considered:

(a) All the data submitted in the application of BPF “C(M)IT/MIT SOLVENT BASED”, as well as the conclusions of the discussions in the BPC and its Working Groups.

(b) Any further information submitted by the applicant or other interested parties in the scope of this request within a timeline specified by the eCA or ECHA as appropriate.

(c) The minority position expressed by Germany and the comments made by Member States during the BPC 34 meeting.

(d) The assessment carried out by Member States in order to ban or limit the use of chlorine compounds in fuel that have legislation in place[6].

(e) The projections for dioxin emissions for road transport for 2005 given in the report “European Dioxin Inventory – Stage II.

(f) The data included in the summary report “Compilation of EU Dioxin Exposure and Health Data”.

Reports mentioned along the exchange with other Member State have been also considered as key information: e.g. the report "Dioxins emissions from Motor Vehicles in Australia" from the Department of the Environment and Heritage of the Australian Government (Smit *et al.,* 2004) or "The European Dioxin Emission Inventory Stage II Volume 2" (Quass *et al*., 2000).

Relevant information from each publication was, as much as possible, summarized. It should be noted that, due to time constraints, publications were not reviewed from a methodological point of view. The process driving dioxin emission resulting from engine combustion is complex and many factors, such as operating conditions, engine speed, fuel and oil composition, after treatment technologies, can affect PCDD/F emissions from vehicles. Moreover, results are highly dependent on analytical methods. It was

[5] EFSA Journal (2010). Application of systematic review methodology to food and feed safety assessments to support decision making. EFSA Guidance 8(6):1637. DOI: 10.2903/j.efsa.2010.1637

[6] Technical information provided by NL CA with regards to national legislation (Besluit Organisch Halogeengehalte Brandstoffen, 1989).

not possible to do an in-depth analysis of all of this. The methodological aspects or experimental designs were not analyzed and no publication was consequently disregarded due to methodological bias.

2. Bibliographic search strategy

1.1. Review related to the formation of dioxins from the use C(M)IT/MIT in fuels

Description of the review method

Period: until 18-09-2020.

Language: no restriction.

Field: abstract/title.

Source: Scopus & PublMED and additional documents listed in annex 1.

Query syntax on Scopus: TITLE-ABS-KEY(Méthylisothiazolinone OR CMIT* OR MIT OR "5-chloro-2-methyl-2H-isothiazol-3-one" OR "2-methyl-2H-isothiazol-3-one" OR "26172-55-4" OR "55965-84-9" OR "2682-20-4") AND TITLE-ABS-KEY("polychlorinated dibenzodioxins" OR "polychlorinated furans" OR PCDD* OR PCDF OR dioxin*)

Query syntax on PubMED : (Méthylisothiazolinone[Title/Abstract] OR CMIT*[Title/Abstract] OR MIT[Title/Abstract] OR "5-chloro-2-methyl-2H-isothiazol-3-one"[Title/Abstract] OR "2-methyl-2H-isothiazol-3-one"[Title/Abstract] OR "26172-55-4"[Title/Abstract] OR "55965-84-9"[Title/Abstract] OR "2682-20-4"[Title/Abstract]) AND ("polychlorinated dibenzodioxins"[Title/Abstract] OR "polychlorinated furans"[Title/Abstract] OR PCDD*[Title/Abstract] OR PCDF[Title/Abstract] OR dioxin*[Title/Abstract])

Eligibility criteria for inclusion or exclusion of studies

Any study giving an estimation of the amount of formation of dioxins due to the use of "C(M)IT/MIT SOLVENT BASED" in fuels has been considered. Exclusion was based on titles and abstracts analysis.

Output of the articles search

The query returns 68 document results but none concern C(M)IT/MIT used in fuels. Most of them are returned since "MIT" (i.e. "with") appears in the German translation of the abstract. Some refers to the use of Isothiazolinones in cleaning products or shampoo matrices.

1.2. Review related to the formation of dioxins from road traffic and the impact of adding chlorine compounds in fuel on dioxin emission

Description of the review method

Period: until 18-09-2020.

Language: no restriction.

Region: exclusion of South and Central America, Africa, some Asiatic countries (see below), Russian Federation and neighbouring countries (e.g. Ukraine, Estonia). Some publications (especially from Asia) have been reintegrated by subsequent search following snowball method.

Field: abstract/title.

Source: Scopus and additional documents listed in annex 1.

Champ: abstract/title keywords.

Source: Scopus.

Query syntax on Scopus :

TITLE-ABS-KEY (fuel* OR petrol) AND TITLE-ABS-KEY (car* OR transport OR traffic) AND TITLE-ABS-KEY ("polychlorinated dibenzodioxins" OR "polychlorinated furans" OR pcdd* OR pcdf OR dioxin*) AND (EXCLUDE (AFFILCOUNTRY , "Russian Federation") OR EXCLUDE (AFFILCOUNTRY , "India") OR EXCLUDE (AFFILCOUNTRY , "Mexico") OR EXCLUDE (AFFILCOUNTRY , "Pakistan") OR EXCLUDE (AFFILCOUNTRY , "Turkey") OR EXCLUDE (AFFILCOUNTRY , "Ukraine") OR EXCLUDE (AFFILCOUNTRY , "Estonia") OR EXCLUDE (AFFILCOUNTRY , "Hong Kong") OR EXCLUDE (AFFILCOUNTRY , "Malaysia") OR EXCLUDE (AFFILCOUNTRY , "Cambodia") OR EXCLUDE (AFFILCOUNTRY , "Colombia") OR EXCLUDE (AFFILCOUNTRY , "Egypt") OR EXCLUDE (AFFILCOUNTRY , "Jordan") OR EXCLUDE (AFFILCOUNTRY , "New Zealand") OR EXCLUDE (AFFILCOUNTRY , "Puerto Rico") OR EXCLUDE (AFFILCOUNTRY , "South Africa") OR EXCLUDE (AFFILCOUNTRY , "Tunisia") OR EXCLUDE (AFFILCOUNTRY , "United Arab Emirates") OR EXCLUDE (AFFILCOUNTRY , "Viet Nam") OR EXCLUDE (AFFILCOUNTRY , "Zimbabwe")) AND (EXCLUDE (SUBJAREA , "MEDI") OR EXCLUDE (SUBJAREA , "PHYS")) AND (EXCLUDE (SUBJAREA , "EART") OR EXCLUDE (SUBJAREA , "PHAR"))

Principles for inclusion or exclusion of studies

At the screening step, exclusion was based on title and abstract analysis. Only study providing information on (i) mechanism of dioxin formation (e.g. reviews), or on (ii) estimation of the amount of dioxins formation due to vehicles/road traffic and of its contribution to total emission (from all sources) was considered. The others were disregarded. Many of the publications retrieved initially did not contain information relevant for the specific purposes of this study (e.g. publications reported on health impacts of (diesel) exhaust).

At eligibility step, publications were briefly reviewed (including results and discussion) and only the more relevant ones (i.e. reviews, inventories and most cited publications) were taken into account.

Output of the articles search

As a result, a total of 307 publications were identified to be potentially interesting for this study. Around thirty articles were finally selected to draw up this overview on the formation of dioxins due to road traffic.

Publications related specifically to dioxin emission due to water transport were not identified and this issue was therefore not considered in this work.

IV. Bibliographic synthesis

1.1. The formation of dioxins from the use C(M)IT/MIT in fuels

As explained in the chapter III.1.1, no publication concerning C(M)IT/MIT used in fuels and the formation of dioxins was retrieved from the search. It was therefore not possible to answer the questions raised by the EU Commission on the amount of formation of dioxins from the use C(M)IT/MIT in fuels and its contribution to total emissions of dioxins, as well as, on the risks associated with this use for human health and for the environment.

1.2. The formation of dioxins from road traffic and the impact of adding chlorine compounds in fuel on dioxin emission

a. The mechanism dioxin formation in vehicle engines and exhaust systems

The formation of dioxins through general combustion processes has been thoroughly described in the scientific peer reviewed literature, especially in the case of waste incineration systems (Bremmer *et al*., 1994; Huang & Buekens, 1996; Stanmore 2004; Cheruiyot *et al*., 2016). In the specific case of internal combustion engines, the process is considered similar, i.e. driven by the chemistry of incomplete combustion and not affected by the technical performance of the combustion process (Smit *et al.,* 2004; Broz *et al*., 2000).

The formation of dioxins could proceed either in the vapour phase (homogeneous reaction), or on solid surfaces such as soot or ash particles (heterogeneous catalysed reaction). For both types of reaction, the usual residence time of gas and solids is of the order of seconds but the critical temperature ranges of 800 to 500 ◦C in the first case (gas) and 400 to 200 ◦C (solid) in the second one (Stanmore, 2004; Cheruiyot *et al*., 2016).

As for incinerators, the formation of dioxins could result from *de novo* reaction (oxidation and chlorination of any unburned carbon in the particulates) for diesel (Jones, 1993) as well as petrol engines (Stanmore, 2004). It takes place in the low-temperature post-combustion region (exhaust system) where the active agents are oxygen and chlorine supplied from the solid itself and/or the gas phase (Huang & Buekens, 1996; Stanmore, 2004).

Dioxins are formed when four important process conditions are met:

(1) temperature between 200-800 °C (ideal range between 200-400 °C);

(2) presence of chlorine;

(3) presence of organic (specifically aromatic) matter;

(4) presence of oxygen (Bremmer *et al*., 1994; Stanmore, 2004).

For heterogeneous reaction, particle concentrations (e.g. fly ash), which is influenced by the extent of gas cleaning (Stanmore 2004) and the presence of metals (which serve as catalysts), affect also strongly the formation of dioxins.

Overall, there is no doubt from literature that **motor vehicles powered with fuels containing chlorine are potential sources of dioxin and furan emission, however the magnitude of their total input remains uncertain** (Smit *et al.,* 2004; Laroo *et al*. 2011; Cheruiyot *et al*., 2016).

b. Estimation of PCDD/PCDF emission variability

Emission factors correspond to the total amount of dioxins emitted per liter of fuel, usually expressed as pg/L of fuel or pg I-TEQ/ L of fuel[7]. In the last case, the quantities of dioxins are given by the sum of individual amounts of dioxin-like congeners in the mixture, expressed as 2,3,7,8-TCDD toxic equivalents quantity (I-TEQs). This expression allows to weigh the amount of dioxin-like congeners with regard to their level of toxicity compared to the toxicity of the most highly studied dibenzo-p-dioxin, 2,3,7,8-TCDD. These values need to be combined with the fuel economy data for each vehicle type in order to obtain emission factors expressed as pg I-TEQ /km, which are required to estimate the contribution of traffic to total dioxin emission.

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7 International scheme ITEQ has been proposed by the North Atlantic Treaty Organisation (NATO): NATO/CCMS, 1988.

Published emission factors for road traffic show very large variability (by over a factor of 200: Geueke *et al*., 1999; Cheruiyot et al., 2016), even for the same car under comparable test conditions (Broz *et al*., 2000). This can be attributed to many factors: measurement methods and approach (laboratory/tunnel/on road; particulate/gaseous phase; detection limits…), operating conditions (engine speed rate cycle), compounds identity and toxic equivalence pattern, fuel and oil characteristics (especially scavenger content), engine characteristics.

Table 1 presents a summary of values reported in the review published by Srogi (2008), Dopico & Gómez (2015) and Smit *et al. (*2004). When possible, values are differentiated according to country, experiment type, vehicles technology type and fuel type. In these reviews, details (e.g non-catalyst passenger cars, catalyst passenger cars, light-duty vehicles, heavy-duty vehicles or fleet) are however not available for all studies.

Table 1: **Dioxin emission factors from vehicles in different countries**

Study and year	Country	Sampling	Vehicles	EF* (pg I-TEQ/km)	EF2 (pg I-TEQ/L)	Cl (ppm)
Hagenmaier *et al.*, 1990	Europe (Germany)	Tailpipe	Unleaded gasoline Unleaded gasoline Leaded gasoline Diesel Diesel***	5.1 0.7 108.3 2.4 35***	50.7 7.2 1083 23.6 70***	<1 – 48 – –
Buhler & Greiner, 1996 ***	Europe (Germany)	Tailpipe	Diesel	14	77	–
Oehme *et al.*, 1991	Europe (Norway)	Tunnel	Leaded /unlead G.LDV HDDV Diesel	38–520 720–9500	– –	–
Wevers *et al.*, 1992***	Europe (Belgium)	Tunnel	Leaded gasoline Unleaded gasoline Diesel	1641 10 35.7	– – –	– – –
Eduljee & Dyke, 1996**	Europe (UK)	-	Leaded gasoline	1,1-220	–	–
			Diesel	0.65-10	–	–
			Unleaded with catalytic converter	0.36-13	–	–
			Unleaded without catalytic converter	0.36-21	–	–
Pacyna *et al.*, 2003 **	Europe	-	Leaded gasoline	0.5-270	–	–
		-	Unleaded gasoline	0.104-10	–	–
Marklund *et al.*, 1987	Europe (Sweden)	Tailpipe	Leaded gasoline	30-540	–	–
Marklund *et al.*, 1990	Europe (Sweden)	Tailpipe	Unleaded gasoline LDV Leaded gasoline LDV Diesel	0.36 1.1-2.6 nd	3.5 10-23 nd	14 63 0.61
Chuang *et al.* 2010 ****	Asia	Tailpipe	Motorcycle gasoline	86-114	–	–
Chuang *et al.* 2011	Asia	Tailpipe	SUV unleaded gasoline	123	–	–
			DPV Diesel	80	–	–
			HDDV Diesel	1690	–	–
Chang *et al.*, 2004	Asia	Tunnel	Unleaded gasoline Diesel	22.9 91.7	229.3 550.4	–
Gullet & Ryan 2002	USA	On road Tailpipe	New HDDV Diesel	8-23		
		On road Tailpipe	Old HDDV Diesel	164		
Ryan & Gullet, 2000 ***	USA	On road Tailpipe	HDDV Diesel	29–106	–	< 20
Lew, 1993 & 1996 ****	USA	Tailpipe	HDDV Diesel	663–1300	–	–
Gertler *et al.*, 1998 ****	USA	Tunnel	Diesel	170 ± 80	–	–
CARB, 1987***	USA	Tailpipe	Leaded gasoline LDV	203	1794	
			Diesel	380	5904	
			Diesel Truck	4900	27440	

Laroo et al., 2012			Diesel HDDVs		7.11–13.6	9.8
Bingham *et al.*, 1989***	New Zealand	Tailpipe	Leaded gasoline	15–39		123

HDDV: Heavy Duty Vehicle; SUV: Sports Utility Vehicle; DPV: Diesel Passenger Vehicle; LDV: Light Duty Vehicle

* Equivalent factors

** Estimations from literature. Eduljee & Dyke 1996: estimations based on Eadon-TEQ (attention column headings are staggered).

*** Cited by Srogi (2008). For Ryan & Gullet (2000), the values correspond to the average and upper 95% CL for whole dataset (i.e. tests conducted for two distinct driving routes). Lower and higher values were obtained for the city test: 3-96 pg I-TEQ/km.

**** Cited by Cheruiyot et al. (2016).

As example, Smit et al. (2004) has summarized for Australia the following range to estimate the national PCDD/PCDF emission factor (calculated with the national fuel economy data).

Petrol vehicles emission

1. non-catalyst passenger cars running on leaded petrol: 10-140 pg I-TEQ/km
2. non-catalyst passenger cars running on unleaded petrol: 2-20 pg I-TEQ/km
3. catalyst passenger cars running on unleaded petrol: 1-3 pg I-TEQ/km.

Diesel vehicles emission

1. light-duty vehicles running on diesel (passenger cars, LCVs): 6-50 pg ITEQ/km
2. heavy-duty vehicles running on diesel (trucks): 15-104 pg I-TEQ/km
3. buses running on diesel: 12-85 pg I-TEQ/km.

These values are not strictly comparable (in particular due to methodological differences) but give the range of the main differences between fuel types (leaded petrol, unleaded petrol, diesel) and vehicle technology types (e.g. non-catalyst passenger cars, catalyst passenger cars, light-duty vehicles, heavy-duty vehicles). Overall data show higher emissions for leaded petrol, which are mainly attributed to the “scavenger-compounds” added to the fuels to prevent the engine from the formation of lead precipitates (Marklund *et al*., 1987; 1990; EC, 2000). The level of emission from diesel is less clear and its potential contribution to total dioxin emissions is still controversial. High emissions were reported in the US and Europe, especially for heavy-duty diesel vehicles, with large variability in congener profiles (Oehme *et al.*, 1991; Jones, 1995; Clunies-Ross *et al*.; 1996; Gertler *et al*.; 1998; Laroo *et al.,* 2011). But several studies have reported low emissions -if not zero- for various diesel vehicles, at amounts comparable to unleaded gasolines (Hagenmaier, 1994; Geueke 1999; EC, 2000). This contradictory result could be partly due to differences in measurement methods (test rig / tunnel).

It is worth to note that all these values are representative of various cases throughout the world, from eighties to 2010 but would need to be updated to describe more precisely the potential emission from current fleet and fuel uses in Europe.

c. Impact of adding chlorine compounds in fuel on dioxin emission

Chlorine is a key precursor of the dioxin formation from combustion (US EPA, 2000). During *de novo* formation, it is supplied from solid (e.g., by the decomposition of a metal chloride), or by the chlorinated products of incomplete combustion, or the chlorine present in the gas phase (Stanmore 2004). Although well studied, the link between fuel chlorine content and the level of dioxin formation is however far from simple (Dyke *et al*., 2007; Rey *et al*., 2014; Wang *et al*., 2012). Strong correlations between chlorinated additives in petrol and engine oil and PCDD/PCDF emissions were shown (e.g. Marklund *et al*., 1987; Hagenmaier *et al.,* 1990; Bacher *et al*., 1991). Similarly, reduction of PCDD/PCDF and PCB emissions was reported when halogens, especially chlorine, are excluded from fuels and oils (Broz *et al*., 2000). This relationship was presented as the main explanation of the high dioxin emission from leaded petrol (characterized by higher chlorine content, 50-300 ppm while unleaded petrol content is 1- 10 ppm, Smit *et al.,* 2004). Such high contents are largely due to the addition of halogenated additives or 'scavengers' to leaded petrol or to engine oil, used to clean the engine by conversion of lead deposits into volatile compounds. The use of chlorinated additives in fuels was therefore suspected to be a key factor driving dioxin formation in vehicle engines, leading to regulate their content in fuels in some countries. This view is however not supported by other authors (Dyke *et al*., 2007).

Linear relationships between chlorine content and dioxin formation has indeed not been observed as a rule for the various fuel and vehicle technology types. Several experiments show that chlorine content has no or nonlinear impact on the synthesis of PCDD/Fs (Dyke *et al.* 2007; Liu *et al*. 2011; Chang *et al*., 2014) while others have shown that fuel containing chlorine, especially when catalytic conditions with copper occur, can significantly increase the formation of PCDD/Fs for diesel fuels (Heeb *et al.* 2007; Mayer *et al*., 2003). According to several authors, it seems that the influence of chlorine on the formation mechanisms of PCDD/Fs depends on its content in fuels (and wastes): relationship exists only when concentrations of chlorine in the feed[8] /fuels are small (less than or near 1%) and when poor combustion occurs (Smit *et al*., 2000; Stanmore, 2004; Wang *et al*., 2003). Besides other sources of chlorides (e.g. inorganic chlorides in the atmosphere: Broz *et al*., 2000) could influence the release of dioxin and complicate the interpretation of results. Finally, reliable description of the relationship between fuel chlorine content and dioxin emission from fuel combustion (i.e. mathematical model with validity domains) cannot be derived from scientific literature.

Hence, estimation of dioxin emissions cannot be derived from maximum level of chlorine contribution to the fuel from C(M)IT/MIT and conversion yield of chlorine calculated from publications as proposed by the applicant (chapter VI).

d. Contribution of traffic to total dioxin emission from all sources

In most – if not all – dioxin emission inventories, once emission factors (I-TEQ/km) were established, estimations of the total vehicle kilometers travelled per year are used to estimate the contribution of traffic to total dioxin emission at a national scale.

The Table 2 summarizes the contribution of traffic to total dioxin emission found in the literature. Overall, dioxin emissions from road traffic were estimated to be in the range of 0.2 % to 12.3 % with most of estimations under 3 %. Stationary combustion sources (e.g municipal solid waste incinerators, metal production processes and industrial incinerator) are considered as the main contribution of dioxin emissions (CE, 2000; Caserini & Monguzzi, 2002; Cheruiyot *et al*., 2016). As shown in table 2, situations are clearly very different from one country to another. This discrepancy is not only due to differences of the specificities of countries in terms of road traffic or industrial equipment but also probably to the differences between methods followed to obtain these estimations and to the uncertainty surrounding each individual estimation of dioxin emission. Random and systematic errors (bias) in parameter datasets (emission factors, fuel economy data, distance travelled per year) used in such inventories lead to multiply

[8] Part of this knowledge was established from studies on incinerators, where feed refers to the material, mainly wastes, used in the combustion chamber

the uncertainty of the final estimations. Such global emission estimations and the low contribution of road traffic should therefore be interpreted with precaution.

Table 2: Dioxin emissions from road traffic in various emission inventories (g ITEQ/year)

Reference Report/Paper	Year	Country	Dioxins Total [g I-TEQ/year]	Dioxins from road Traffic [g I-TEQ/year]	Dioxins from road Traffic [% of Total]
Eduljee & Dyke, 1996	1992	UK	560-1100	1-45	0.2-4.1
Hagenmaier et al., 1994*	1994	Germany	800-1200	11-50	1.38-4.17
Bremmer *et al.*, 1994	1991	The Netherlands	484	7	1.45
Bremmer, 1997	1997	The Netherlands	58	0.8	1.38
EC, 1997*	1997	EU	3276	19	0.58
Quass *et al.*, 2000**	1995	EU	6470	138	2.13
	2000		4660	82	1.76
Geueke *et al.*, 1999	1999	EU	6000	31.5	0.53
Caserini & Monguzzi, 2002	1997	Italy (Lombardy)	33	3.86	11.7
US EPA, 2000	1987	USA	12 331	61.5	0.50
	1995	USA	2 888	41	1.41
US EPA, 2006***	2000	USA	1 314	-	4.7 (diesel heavy duty trucks)
Miyabara *et al.*, 1999	1994	Japan	5000	17	0.34
Environment Canada	1997	Canada	290	9	3.00
Xuan *et al.*, 2017	-	China (Suzhou City)	-	-	12.3
Taiwan EPA, 2008***	2008	Taiwan	74.7	2.31	3.1
Bawden, 2004	2000	Australia	500	-	1 (diesel engines)*

* Cited by Smit *et al.* (2004).

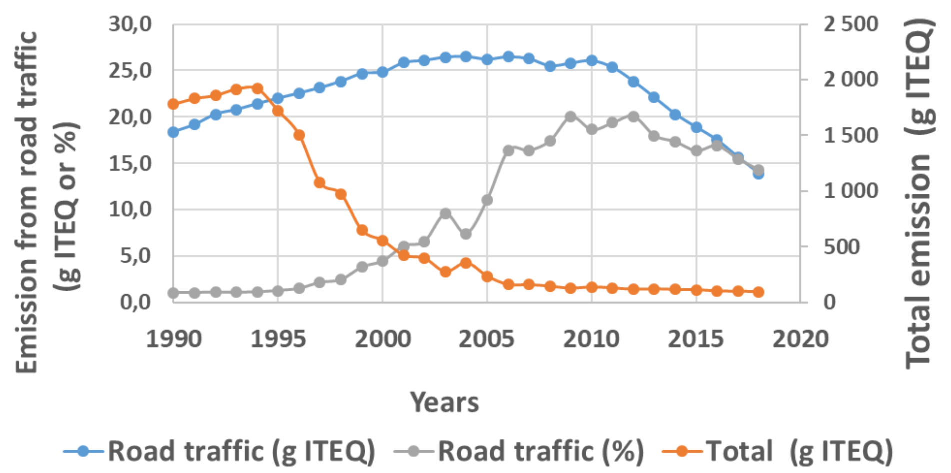
** Maximum estimates for emissions from road traffic compared with upper total emission estimate.

*** Cited by Cheruiyot *et al.* (2016) and Wang *et al.* (2010).

According to the European Dioxin Inventory (Quass *et al.*, 2000), PCDD/F emissions from road traffic have experienced a significant decrease of 39% of dioxin emissions from road transport between 1995 and 2000 (preceding another expected decrease estimated around 48% between 2000 and 2005). This evolution is mainly attributed to the reduction of the lead content in petrol and the decrease of the use of scavengers. In the meantime, however, higher reductions were observed for dominant industrial sources, e.g. municipal and industrial waste incinerators, due to the implementation of new regulation and technical progress (Quass *et al.*, 2000 ; Dopico & Gómez, 2015). Thus, although still minor compared to current dominant sources, the relative proportion of non-industrial diffuse sources, including transport, in total emissions is likely to increase (EC, 2000; Chen 2004; Dopico & Gómez, 2015). The last reviews on this topic (Quass *et al.*, 2004; Dopico & Gómez 2015) conclude that the role of non-industrial diffuse sources, including transport, is becoming more and more important and might be, in the future, the overall main contributor in the formation of dioxins.

The last national inventory on air pollution in France (Serveau *et al.*, 2015) gives a good illustration of the increase of the contribution of road traffic to total dioxins emissions (figure 1 including updated data taken from CITEPA[9] website). In 2018, the total emission was estimated at 13.9 g I-TEQ (i.e. 14.3 % of a total estimated at 97 g ITEQ). The significant increase of this contribution (from 1% to 14.3 %) is mainly due to the noteworthy decrease of the total emissions of dioxin (95 % from 1990 to 2018 according to Serveau *et al*., 2015). A more accurate estimation was made for the urban area of Lyon where the contribution rose from 0.1% to 3.7 % of total dioxins emissions (Coudon, 2018). Dioxin emissions were over there mainly attributed to industry in 1991, 1996 and 2002, then, to diffuse sources between 2002 and 2008, (56% of total dioxin emissions).

Figure 1: Estimations of dioxin emissions from road traffic and its contribution to total emission from 1990 to 2018 in France (from https://www.citepa.org)



Since leaded fuel have been faded out in Europe, the remaining PCDD/F emissions from traffic is now considered to depend mainly on the extent of diesel combustion (EC, 2000; Smit et al. 2004). This has already been noted for Lombardy region, where the contribution of diesel vehicles was estimated to be the main part of traffic emission (77.6 % of the total 3.863 g I-TEQ/year; Caserini & Monguzzi, 2002). The rapid evolution of this industrial sector with the arrival of new fuels, engines and after-treatment technologies raises however new questions. These last years, the debate indeed has moved toward the potential role of after-treatment technology (used to reduce pollutant, i.e. catalytic pipe) and new fuels (biofuels or fuel doped with the addition of organic metal compounds such as copper). Here too, results do not allow drawing simple meaningful conclusions. Some studies conclude that doped fuels and exhaust after-treatments can reduce significantly PCDD/F emissions (Bremmer, 1994; Laroo *et al*., 2011; Liu *et al*., 2011) but others have shown the opposite (Clunies-Ross *et al*., 1996; Chuang *et al*., 2011; Heeb *et al*., 2015; Mayer *et al*., 2003). After-treatment does not appear, for the moment, as the absolute solution to limit dioxin emission from traffic. This rapid evolution raises the question of the relevance of some of the datasets used in the inventories presented above.

Although emissions from traffic are minor compared to current dominant sources, it should be noticed that their relative importance in terms of relative human exposure could be higher, due to their spatial distribution in densely populated urban areas, close to the human population and living environment and with less dilution of emissions (Smit *et al*., 2004; Cheruiyot *et al*., 2016). Besides, additional specific contamination pathway was observed for food transported by road, which could contribute to the total exposure of population (Marklund *et al.,* 1990).

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[9] CITEPA (Centre Interprofessionnel Technique d'Etudes de la pollution atmosphérique): non-profit organisation and State operator for the French Environment Ministry.

As a conclusion, it should be noted that:

- **Inventories state that dioxin emissions from road traffic are minor – few percent - in comparison with other quantified sources but its relative contribution to total emission could have been increase these last years due to the reduction of industrial emissions.**
- **All these estimations are characterized by very large uncertainties and much of the data used to estimate road traffic emissions in these inventories date back 15-20 years. Their relevance to describe current situation shall be investigated.**
- **As it is impossible to assess the contribution of the use of "C(M)IT/MIT SOLVENT BASED" in fuels to dioxin formation, this review has been focused on dioxin emissions from road traffic whatever their origin. It should be noted however that the use of C(M)IT/MIT in fuel is not restricted to automotive fuels but can includes also fuels for residential heating. Such use shall be considered in the estimation of the potential amount of formation of dioxins resulting from the use of "C(M)IT/MIT SOLVENT BASED" in fuels.**

V. Theoretical calculation on the potential formation of dioxins from the combustion of C(M)IT/MIT in fuels

a. Calculation from Ctgb

In response to the request from ECHA (SECR) on the potential of national legislation related to the formation of dioxins due to the addition of chlorine- or bromine containing substances to fuels, Ctgb[10] has indicated, the existence for NL of the following legislation -Besluit Organisch Halogeengehalte Brandstoffen (BOHB, 1989)- which states that:

1. It is not allowed to use fuels:
   - if it contains polychlorinated biphenyl exceeding 0,5 mg/kg per congener, or
   - if it contains organic halogenic compounds exceeding 50 mg/kg.
2. Notwithstanding section one, part b, airplane fuels can be used with a concentrations of organic halogenic compounds exceeding 50 mg/kg, but no more than 500 mg/kg.
3. It is not allowed to use organic halogenic compounds, or preparations exceeding the concentrations stated in section 1 or 2 in the manufacturing of fuels. Manufacturing of fuels, or the substances or preparations referred to, includes mixtures of these substances or preparations of fuels.

In this perspective, Ctgb has provided a calculation in order to check if the dosage as prescribed in the SPC of the biocidal product family "C(M)IT/MIT SOLVENT BASED" can lead to unacceptable concentrations of PCBs and dioxins in fuels, according to this legislation (Ctgb, 2020[11] - Appendix 1). This theoretical calculation assumes a total conversion of moles of chlorine atoms from C(M)IT/MIT into the most toxic dioxine 2,3,7,8-TCDD (2,3,7,8-tetrachlorodibenzo-p-dioxin) (used as a reference in the international toxic equivalence factor ITEQ scheme). By this way, an estimation of [redacted] was obtained. Similarly, a theoretical estimation of the potential formation of PCB was provided.

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[10] College voor de toelating van gewasbeschermingsmiddelen en biociden.
[11] Appendix 1 - Ctgb (2020). Technical information provided by NL CA with regards to national legislation.

It was concluded from these theoretical calculations that the dosage as prescribed in the SPC may lead to concentrations of PCBs and dioxins in fuels above NL legal thresholds.

This calculation does not allow to estimate the amount of dioxins formed due to the use of "C(M)IT/MIT SOLVENT BASED" in fuels.

b. Theoretical calculation of dioxin emissions from the applicant

Theoretical calculation of dioxin emissions derived from the use of the products of C(M)IT/MIT SOLVENT-BASED family for preservation of fuel has been proposed by the applicant (Applicant position paper, 2020[12] - Appendix 2; Applicant position paper, 2021[13] - Appendix 3).

This theoretical calculation assumes conversion of chlorine atoms[redacted], into dioxin, by considering:

1. Maximum level of chlorine contribution to the fuel [redacted] at the application rate proposed for authorization (3-6 ppm of C(M)IT/MIT for curative treatment).
2. [redacted]
3. Estimation of the conversion yield of chlorine into TCDD equivalents (TEQ) calculated from the study of Marklund *et al.* (1990). The applicant has indicated that, according to his analysis of the scientific literature, the value appears as a reasonable estimate for essentially all motor classes except diesel engines with copper-catalyzed particulate traps.
4. Estimation of TEQs emissions from automotive fuel preserved with CMIT/MIT considering the estimated conversion yield.
5. An annual tonnage in Europe of[redacted]CMIT/MIT SOLVENT-BASED product [redacted]
6. A worst-case bulk density of 0.75 kg/L for both fuel types required for conversion from annual fuel tonnage to volume.

Total dioxin emission was finally estimated by calculating the yearly total volume of fuel necessary for a treatment with the whole annual tonnage of C(M)IT/MIT solvent, at the dose considered for curative treatment (3 -6 ppm).

This approach results in a total maximum emission of[redacted] in the whole of Europe from the use of the C(M)IT/MIT solvent as preservative in crude oil and middle distillate fuel. This estimation is deemed by the applicant as relevant for essentially all motor classes except diesel engines with copper-catalyzed particulate traps.

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[12] Appendix 2. Position paper applicant submitted after BPC (2020). Use of products of the CMIT/MIT solvent-based biocidal product family for preservation of fuel for automotive use – Addressing concerns related to potential formation of dioxins during combustion.

[13] Appendix 3. Position paper applicant submitted after BPC (2021). Comments to the response to the EU Commission mandate requesting ECHA opinions under Article 75(1)(g) of the BPR "Estimation of the emissions of dioxins from the use of the "CMIT-MIT SOLVENT BASED" biocidal product family (BPF) as preservative in crude oil and middle distillate fuel"

These calculations present several shortcomings.

1. It assumes a linear relationship between chlorine content and dioxin formation, while the review of the scientific literature (chapter 1.3) has shown that such relationship has not been observed as a rule for the various fuel and vehicle technology types.

2. The choice of the conversion yield calculated from study of Marklund *et al.* (1990) with regard to total Cl content in fuel and I-TEQ dioxin emissions is not justified by any analysis of the scientific literature.

   It is hardly possible from the scientific literature to derive a reasonable worst case conversion yield representative of current EU fuels and vehicle technology types. To this end, we would need a clear representation of the relationships between chlorine content (entering the combustion chamber) and the dioxin formation, i.e. equations, parameters and domain of validity clearly defined. Instead we have no consensus on the relevance of such a relationship (as general rule), very few couple of values for chlorine content / dioxin emission available in the literature, and numerous studies representatives of outdated fuels/engines and partly specific to non EU countries (see table 1).

   Moreover it is worth to note that the I-TEQ dioxin emission (expressed as pg I-TEQ/l or pg I-TEQ/km) reported by Marklund *et al.* (1990) does not appear as worst-case compared to the values listed in Table 1. Anyway, to determine the worst case published data, it is necessary here to compare conversion yield of chlorine into TCDD equivalents (so to know the chlorine content of fuel), not only dioxin emission values.

   Lastly but not least, it is besides not clear from which values from the Marklund *et al.* (1990), the conversion yields conversion of chlorine into TEQs has been estimated (no explanation in the position paper)[14]. The values are expressed in % ($7.9.10^{-8}$ and $9.5.\ 10^{-8}$ %) while the values taken for the calculations are expressed in pg I-TEQ/l (dioxin emission) and in ppm (fuel chlorine content).

3. [redacted]

4. The calculation of [redacted] input is based on a treatment of 6 ppm v/v of KATHON FP while the appropriate value would be 400 ppm (6 ppm is the content of CMIT/MIT for curative treatment).

It is concluded that due to these shortcomings the calculation does not allow to estimate the amount of dioxins formed due to the use of “C(M)IT/MIT SOLVENT BASED” in fuels.

c. Theoretical calculation of dioxin emissions

A worst-case theoretical estimation of the dioxin emissions resulting from the use claimed for the products C(M)IT/MIT solvent-based is proposed below. The approach followed for this calculation is quite similar to that proposed by Ctgb combined with the suggestion proposed by the applicant on the estimation of the yearly total emission in Europe from the yearly total volume of C(M)IT/MIT used in CMIT/MIT SOLVENT-BASED . It is assumed that:

- a total conversion of moles of chlorine atoms into dioxin or PCB. As CMIT contains 1 chlorine atom, 1 mole C(M)IT/MIT (3:1) yields 3 moles of chlorine atoms.

[14] Or if the yields are calculated from the values reported in table 2 of the position paper, from where the values of TEQ Emissions (11.2 pg/L for unleaded fuel and 0.55 pg/L for Diesel fuel) are coming from.

- each chlorine atom from the product allows the formation of the most toxic dioxin is 2,3,7,8-TCDD (2,3,7,8-tetrachlorodibenzo-p-dioxin (used as a reference in the international toxic equivalence factor ITEQ scheme).
- [redacted].

Worst case estimation of dioxins formation per liter of fuel

The following parameters were considered:

- Molar weight C(M)IT/MIT = 564 g/mol (CMITMIT Mw)
- Molar weight Cl = 35.5 g/mol (Cl Mw)
- Max content C(M)IT/MIT in fuel, taking the purity in TC/TK into account (at the maximal application rate for curative treatment = 7 ppm[15]) (Dose CMITMIT)
- Bulk density of C(M)IT/MIT (pure) = 1547.5 g/L (ρ_1)
- Bulk density of the product [redacted] = 1057,6 g/L (ρ_2)[16]
- Molar weight 2,3,7,8-TCDD (2,3,7,8-tetrachlorodibenzo-p-dioxin = 321.97 g (Dioxin Mw)
- Content Cl [redacted] in TK material = [redacted] wt (Content Cl)
- Max content of TK material in product [redacted] = [redacted] (Content TK)
- Dose product in fuel (maximal application rate) = 400 ppm v/v (Dose product in fuel)

The dioxin emission resulting from the input of chlorine brought by C(M)IT/MIT (Content Dioxin 1 in mg TEQ/L fuel) is estimated by the following equations:

$$\text{Content Dioxin 1} = \frac{(\text{Dose CMITMIT}) * \rho 1 * 10^{-6}}{(\text{CMITMIT Mw})} * \frac{3 * (\text{Dioxin Mw}) * 10^{3}}{4}$$

The dioxin emission resulting from the input of chlorine brought by [redacted] (Content Dioxin 2 in mg TEQ/L fuel) is estimated by the following equation:

$$\text{Content Dioxin 2} = \frac{(\text{Content Cl}) * (\text{Content TK}) * 10^{-4} * \rho 2 * (\text{Dose product in fuel}) * 10^{-6}}{(\text{Cl Mw})} * \frac{(\text{Dioxin Mw}) * 10^{3}}{4}$$

The total of dioxin emission thus is estimated to be [redacted] mg/L fuel ([redacted]). This calculation corresponds to a worst case scenario where all chlorine atoms are converted into 2,3,7,8-TCDD. [redacted].

Worst case estimation of total dioxin emission from fuel combustion

Two approaches could be followed to estimate total dioxin emission per year:

(1) from fuel economy and traffic data as commonly done in inventories (chapter IV.1.2.c)

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15 Calculated from the rate for curative use 400 ppm of biocidal product x the maximal content of C(M)IT/MIT in the product [redacted]

16 [redacted].

(2) from C(M)IT/MIT tonnage per year as proposed by the applicant.

The first one is not relevant without validated updated datasets on fuel economy and traffic. Furthermore, it does not include domestic use of fuel containing C(M)IT/MIT solvent-based for heating (heating is a significant source of dioxin in urban area: Dopico & Gomez, 2015).

The second approach could be followed based on the C(M)IT/MIT tonnage per year provided by the applicant [redacted]). Considering a worst-case bulk density of 0.75 kg/L for fuels and a treatment rate of 7 ppm C(M)IT/MIT, the total of dioxin emission is estimated at [redacted] tons / year. Owing to the lack of data, this estimation is clearly overestimated and has no real meaning compared to available estimations of the total dioxin emission in Europe (total emissions for road transport for 2005 given in the "European Dioxin Inventory-Stage II" were 41-60 g TEQ/year, Quass *et al.*, 2000).

In the absence of reliable parameters (i.e. consistent with the scientific literature) to describe the relationship between fuel chlorine content and dioxin emission from fuel combustion, it is unfortunately not possible to provide better estimation of the dioxin emission

VI. Conclusion

During the 34th BPC meeting, the consequence of the presence of C(M)IT/MIT in fuel for automotive use on the formation of dioxins was questioned. In this framework, on July 2020, the EU Commission mandated ECHA under Article 75(1)(g) of the BPR (ARES (2020)3922032 – 24/07/2020) to provide (1) estimation of the amount of formation of dioxins (in mg/year TEQ) due to the use of C(M)IT/MIT solvent based mentioned above and the highest amount of product produced per year according to the authorization and (2) estimation of the overall contribution to the emissions of dioxins coming from this use in the worst–case scenario, and comparison to the emissions of dioxin coming from other sectors or sources.

Additionally, the EU Commission has requested a qualitative or quantitative assessment the level of the risks (i) for human health and (ii) for the environment.

To this purpose, an analysis of the scientific literature on the state of knowledge about the potential link between C(M)IT/MIT in fuels and the formation of dioxins was first performed. Due to the lack of result, the scope of this initial search was enlarged to the role of road traffic in dioxin emission. Theoretical calculations on the potential formation of dioxins that could result from the combustion of the chlorine input by C(M)IT/MIT SOLVENT in fuel were also considered.

Several conclusions can be drawn from this work.

1- **Peer-reviewed publications on the potential of dioxin formation resulting from the use of C(M)IT/MIT as preservative in oil and fuel were not found.**

2- **There is no experimental evidence demonstrating the existence or the absence of formation of dioxin from the use of C(M)IT/MIT as preservative in oil and fuel. This phenomenon is likely to occur, but the extent cannot be estimated.**

3- **Motor vehicles are significant sources of emission of dioxins, but minor compared to current dominant sources. However, along with the decrease of dominant industrial sources of dioxins, the relative proportion of non-industrial diffuse sources, including transport, in total emissions of road traffic is likely to increase, at least in some EU countries.**

4- **Overall, high uncertainties are surrounding estimations of dioxins emissions from traffic due to the actual scarcity of measurements with respect to the numerous key factors involved in the process (e.g. fuel and oil characteristics… as detailed below), difference in the methodology used for estimations and analytical measurements. Besides, the global situation behind this issue is highly dynamic (vehicle fleet, traffic level), leading to make some data partially outdated and final interpretation uncertain.**

5- **The addition of chlorine, through halogenated additives or "scavengers", in the fuel increases dioxin formation under certain conditions (e.g. incomplete combustion as observed in vehicle engines). However, relationship between chlorine content and dioxin formation is not yet clarified and does not seem to be broadly linear.**

6- **In the absence of reliable representation and parameters (i.e. consistent with the scientific literature) to describe the relationship between fuel chlorine content and dioxin emission from fuel combustion, it is not possible to provide a realistic estimation of the dioxin emission resulting from the use of C(M)IT/MIT as preservative in oil and fuel.**

7- **Hence, although the potential consequence of the use of C(M)IT/MIT as preservative in oil and fuel cannot be neglected, it is not possible to draw any conclusions either on the magnitude of the potential contribution of the use of C(M)IT/MIT in fuels with respect to dioxin exposure, nor**

on the potential consequence of chlorine additive such as C(M)IT/MIT in fuels on human health and on environment.

8- The issue was limited to the potential dioxin from traffic, but the use of fuel preserved with C(M)IT/MIT for domestic heating shall also be considered.

9- Although emissions from traffic are minor compared to current dominant sources, their relative importance in terms of relative human exposure could be higher, due to their spatial distribution in densely populated urban areas.

Dioxins can then be considered as potential product from the use of C(M)IT/MIT SOLVENT BASED, with POP, PBT or vPvB properties. According to the BPR requirements, a quantitative risk assessment is generally not appropriate for such substances with POP, PBT or vPvB properties and hazard-based approach should be preferred. Indeed, due to their potential to accumulate in the environment, the unpredictability of the effects of such accumulation in the long-term and the fact that such accumulation is in practice difficult to reverse, a safe threshold, below which dioxin emission into the environment could be considered acceptable for human health and environment, cannot be derived.

Finally, dioxins are likely to be formed from the use of C(M)IT/MIT SOLVENT BASED, but the extent of this formation cannot be estimated from the available information.

In that view, experiments have to be designed according to "realistic and worst-case" European scenarios, in terms of vehicle fleet, fuel use and driving conditions (e.g. cycles representative of urban driving scenarios). To this end, the key choices concerning the experimental design [vehicle technology type (including catalytic devices) and age, fuel and oil composition (including potential additives), ambient temperature, engine calibration practices (speed engine pattern) as well as the type of studies (laboratory engine test bench studies, laboratory dynamometer studies, on-road studies and tunnel studies)], have to be discussed beforehand and decided with experts.

These experimental studies should be submitted to provide factual data demonstrating that the use of CMIT/MIT SOLVENT BASED could be a source of dioxins and to measure to what extent.

References

Bawden K., Ormerod R., Starke G. and K. Zeise (2004). Australian Inventory of Dioxin Emissions, 2004, National Dioxins Program Technical Report No. 3, Australian Government Department of the Environment and Heritage, Canberra.

Bingham A.G., Edmunds C.J., Graham B.W. and M.T. Jones (1989). Determination of PCDDs and PCDFs in car exhaust. Chemosphere 1989; 19:669 –673.

Bremmer H.J., Troost L.M., Kuipers G., De Koning J. and A.A. Sein (1994). Emission of Dioxins in the Netherlands. RIVM, Bilthoven, TNO, Apeldoorn, The Netherlands, Report No. 770501018. Link: https://www.rivm.nl/bibliotheek/rapporten/770501018.pdf).

Broz J., Grabic R., Kilián J., Lojkásek M., Marklund S., Ocelka T., Pekárek V., Pribyl J., Tydlitát V. and Vyska J. (2000). The effect of oils on PAH, PCDD, PCDF and PCB emissions from a spark engine fueled with leaded gasoline, Chemosphere, Vol. 41, pp. 1905-1911.

Buhler U. and R. Greiner (1996). Dioxin Emission der Diesel-fahzeuge und des Ubrigen Strassenverkehrs. Germany: Institut fur Verbrennungsmotoren und Kraftfahrwesen, Universitat Struttgart.

California Air Resources Board (CARB) (1987). Determination of PCDD and PCDF Emissions from Motor Vehicles. California Air Resources Board draft report. Test Report No. C-86-029.

Carvalhaes G.K., Brooks P., Azevedo J.A.T., Azevedo G.C., Machado M.C.S., Quitério S.L., Sousa C. and G. Arbilla (2002). Determination of PCDD/F in total suspended particules in Rio de Janeiro: A preliminary study', Organohalogen Compounds, Vol. 56, pp. 461-463.

Caserini S. and AM. Monguzzi (2002). PCDD/Fs emissions inventory in the Lombardy Region: results and uncertainties. Chemosphere. 2002 Sep; 48(8):779-86. doi: 10.1016/s0045-6535(02)00127-3. PMID: 12222771.

Chang MB., Chang SH., Chen YW. and H. Hsu (2004). Dioxin emission factors for automobiles from tunnel air sampling in Northern Taiwan. Sci Total Environ 325:129–138.

Chang Y.C., Lee W.J., Wang L.C., Yang H.H., Cheng M.T., Lu J.H., Tsai Y.I. and L.H. Young (2014). Effects of waste cooking oil-based biodiesel on the toxic organic pollutant emissions from a diesel engine. Appl. Energy 113: 631–638.

Cheruiyot, N.K., Lee, W.J., Yan, P., Mwangi, J.K., Wang, L.C., Gao, X., Lin, N.H. and Chang-Chien, G.P. (2016). An Overview of PCDD/F Inventories and Emission Factors from Stationary and Mobile Sources: What We Know and What is Missing. Aerosol Air Qual. Res. 16: 2965-2988. https://doi.org/10.4209/aaqr.2016.10.0447

Chuang S.C., Chen S.J., Huang K.L., Wu E.M.Y., Chang-Chien G.P. and Wang, L.C. (2010). Gas/Particle Partitioning of Dioxins in Exhaust Gases from Automobiles. Aerosol Air Qual. Res. 10: 489–496.

Chuang, S.C., Huang, K.L., Chen, S.J., Wang, L.C.,Chang-Chien, G.P. and Tsai, J.H. (2011). PCDD/F emissions from gasoline and diesel fueled vehicles. Sustainable Environ. Res. 21: 29–36.

Clunies-Ross C., Stanmore B.R. and G.J. Millar (1996). Dioxins in diesel exhaust. Nature (1996) 381.

Coudon T., Salizzoni P., Praud D., Danjou A.M.N., Dossus L., Faure E. and B. Fervers (2019). A national inventory of historical dioxin air emissions sources in France, Atmospheric Pollution Research, Volume 10, Issue 4, 2019, Pages 1211-1219, ISSN 1309-1042, https://doi.org/10.1016/j.apr.2019.02.004.

Dopico M. and A. Gómez (2015). Review of the current state and main sources of dioxins around the world. J Air Waste Manag Assoc. 2015 Sep; 65(9):1033-49. doi: 10.1080/10962247.2015.1058869. PMID: 26068294.

Dyke P.H., Sutton M., Wood D. and J. Marshall (2007). Investigations on the effect of chlorine in lubricating oil and the presence of a diesel oxidation catalyst on PCDD/F releases from an internal combustion engine, Chemosphere, Volume 67, Issue 7, 2007, Pages 1275-1286, ISSN 0045-6535, https://doi.org/10.1016/j.chemosphere.2006.12.010

Eduljee GH and Dyke P (1996). An updated inventory of potential PCDD and PCDF emission sources in the UK. Science of the Total Environment 177 303-21.

European Food Safety Authority (2010). Application of systematic review methodology to food and feed safety assessments to support decision making. EFSA Journal 2010; 8(6):1637. [90 pp.]. doi:10.2903/j.efsa.2010.1637.

Gertler A.W., Sagebiel J.C., Dippel W.A. and R.J. Farina (1998). Measurements of dioxin and furan emission actors from heavy-duty diesel vehicles. J. Air Waste Manage. Assoc. 48: 276–278.

Geueke K.J., Gessner A., Quass U., Broker G. and E. Hiester (1999). PCDD/F Emissions from Heavy Duty Vehicle Diesel Engines. Chemosphere 38: 2791–2806.

Gullet B. and J. Ryan. 2002. On-road emissions of PCDDs and PCDFs from heavy duty diesel vehicles. Environ. Sci. Technol. 36:3036–40. doi:10.1021/es011376v

Hagenmaier H., Dawidowsky N., Weberruss U., Hutzinger O., Schwind K.H., Thoma H., Essers U., Bühler U. and Greiner R. (1990). Emission of polyhalogenated dibenzodioxins and dibenzofurans from combustion engines, Dioxin 90, Organohalogen Compounds, Vol. 2, Short Papers 2, pp. 329-334.

Hagenmaier H. (1994). Contributions of diesel-powered vehicles and wood burning to overall PCDD/PCDF emissions, Dioxin 94, 11th International Symposium on Chlorinated Dioxins, PCB and Related Compounds, November 21-25 1994, Kyoto, Japan.

Heeb N. V., Zennegg M., Gujer E., Honegger P., Zeyer K., Gfeller U., Wichser A., Kohler M., Schmid P., Emmenegger L., Ulrich A., Wenger D., Petermann J. L., Czerwinski J., Mosimann, T., Kasper M., Mayer, A. (2007). Secondary effects of catalytic diesel particulate filters: Copper-induced Formation of PCDD/Fs. nvironmental Science and Technology, 2007, 41 (16), 5789-5794. DOI: 10.1021/es062962x.

Heeb NV, Rey MD, Zennegg M, Haag R, Wichser A, Schmid P, Seiler C, Honegger P, Zeyer K, Mohn J, Bürki S, Zimmerli Y, Czerwinski J, Mayer A. (2015). Biofuel-Promoted Polychlorinated Dibenzodioxin/furan Formation in an Iron-Catalyzed Diesel Particle Filter. Environ Sci Technol. 2015 Aug 4;49(15):9273-9. doi: 10.1021/acs.est.5b01094.

Huang H. and A. Buekens (1996). De novo synthesis of polychlorinated dibenzo-p -dioxins and dibenzofurans Proposal of a mechanistic scheme. Science of The Total Environment, Volume 193, Issue 2, 1996, Pages 121-141, ISSN 0048-9697, https://doi.org/10.1016/S0048-9697(96)05330-2.

Huang C., K. Chen, Y. Lai, L. Wang, and G. Chang-Chien (2011). Characterization of atmospheric dry deposition of polychlorinated dibenzo-p-dioxins/dibenzofurans in a rural area of Taiwan. Aerosol Air Qual. Res. 11:448–59. doi:10.4209/aaqr.2011.03.0033

Jones K.H. (1993), Diesel truck emissions, an unrecognized source of PCDD/PCDF exposure in the United States, Risk Analysis, Vol. 13, No. 3, pp. 245-252.

Jones K.H. (1995). Diesel engine emissions and the link to human dioxin exposure. Organohalogen Compounds, Vol. 24, pp. 69-74.

Laroo C.A., Schenk C.R., Charles R., L. Sanchez J., and J. McDonald (2011). Emissions of PCDD/Fs, PCBs, and PAHs from a Modern Diesel Engine Equipped with Catalyzed Emission Control Systems. Environ. Sci. Technol. 2011, 45, 15, 6420–6428, DOI: 10.1021/es104220f

Lew G. (1993). Letter to John Schaum Concerning CARB (1987) Draft Report on CDD/CDFs in Vehicle Exhausts. Sacramento, CA: California Air Resources Board, Monitoring and Laboratory Division. Available for Inspection at: U.S. Environmental Protection Agency, Office of Research and Development, National Center for Environmental Assessment, Washington, D.C.

Lew G. (1996). Letter to G. Schweer (Versar, Inc.) Dated January 11, 1996. Sacramento, CA. State of California Air Resources Board, Engineering and Laboratory Branch. Available for Inspection at: US Environmental Protection Agency, Office of Research and Development. National Center for Environmental Assessment, Washington, DC.

Liu Z.G., Wall J.C., Barge P., Dettmann M.E. and N.A. Ottinger (2011). Investigation of PCDD/F Emissions from Mobile Source Diesel Engines: Impact of Copper Zeolite SCR Catalysts and Exhaust Aftertreatment Configuration. Environ. Sci. Technol. 2011, 45, 7, 2965–2972, DOI: 10.1021/es103933e

Marklund S., Rappe C., Tysklind, M. and Egebäck, K.E. (1987). Identification of polychlorinated dibenzofurans and dioxins in exhausts from cars run on leaded gasoline, Chemosphere, Vol. 16, No. 1, pp. 29-36.

Marklund, S., Andersson, R., Tysklind, M., Rappe, C., Egebäck, K.E., Björkman, E. and Grigoriadis, V. (1990). Emissions of PCDDs and PCDFs in gasoline and diesel fueled cars. Chemosphere 20: 553–561.

Mayer A., Heeb N.; Czerwinski J., Wyser M. (2003). Secondary emissions from catalytic active particle filter systems. SAE, 2003-01-0291.

Miyabara Y, Hashimoto S, Sagai M, Morita M. (1999). PCDDs and PCDFs in vehicle exhaust particles in Japan. Chemosphere 1999; 39:143–50.

National Dioxins Program Technical Report No. 2, Australian Government Department of the Environment and Heritage, Canberra http://www.deh.gov.au/industry/chemicals/dioxins/report-2/dioxinemissionfromroadtraffic.html

NATO/CCMS (1988): International Toxicity Equivalency Factor (I-TEF) Method of Risk Assessment for Complex Mixtures of Dioxins and Related Compounds. Pilot Study on International Information Exchange on Dioxins and Related Compounds, Report Number 176, August 1988, North Atlantic Treaty Organization, Committee on Challenges of Modern Society.

Oehme M., Larssen S. and Brevik E.M. (1991). Emission factors of PCDD and PCDF for road vehicles obtained by a tunnel experiment, Chemosphere, Vol. 23, No. 11-12, pp. 1699-1708.

Pacyna J.M. and Pacyna E.G. (2001). An assessment of global and regional emissions of trace metals to the atmosphere from anthropogenic sources worldwide. Environ. Rev. 9, 269–298.

Pacyna J., Breivik K. Munch J. and J. Fudala (2003). European atmospheric emissions of selected persistent organic pollutants, 1970–1995. Atmos. Environ. 37:s119–31. Doi:10.1016/S1352-310(03)00240-1

Quass U., Fermann M. and G. Bröker (2000). The European Dioxin Emission Inventory Stage II. Volume 2 Desktop studies and case studies. This report was prepared by the North Rhine Westphalia State Environment Agency on behalf of the European Commission, Directorate General for Environment.

Quass, U., Fermann, M., Broker, G. (2004). The European dioxin air emission inventory project - Final results. Chemosphere 54, 1319–1327. https://doi.org/10.1016/S0045-6535(03)00251-0.

Rey M., Font, R. and I. Aracil (2014). PCDD/F emissions from light-duty diesel vehicles operated under highway conditions and a diesel-engine based power generator. J. Hazard. Mater. 278: 116–123.

Ryan J.V. and B.K. Gullett (2000). On-Road Emission Sampling of a Heavy-Duty Diesel Vehicle for Polychlorinated Dibenzo-p-dioxins and Polychlorinated Dibenzofurans. Environmental Science and Technology, 2000, 34 (21), 4483-4489. DOI: 10.1021/es991236

Serveau L., Boutang J., Allemand N., Fontelle J.-P. and J. Vincent (2013). Source CITEPA/format SECTEN – Inventaire des émissions de polluants atmosphériques et de gaz à effet de serre en France séries sectorielles et analyses étendues. CITEPA, Paris.

Smit R.,Zeise K.,Caffin A. and P. Anyon (2004). Dioxins Emissions from Motor Vehicles in Australia. National Dioxins Program. Technical Report No. 2. A consultancy funded by the Australian Government, Department of the Environment and Heritage, Canberra. 60 p.

Srogi K. (2008). Levels and congener distributions of PCDDs, PCDFs and dioxin-like PCBs in environmental and human samples: a review. Environ Chem Lett 6, 1–28 (2008). https://doi.org/10.1007/s10311-007-0105-2

Stanmore B.R. (2004). The formation of dioxins in combustion systems. Combust. Flame 136: 398–427.

US EPA (2000). Draft Dioxin Reassessment, United States Environmental Protection Agency, Office of Research and Development. http://cfpub.epa.gov/ncea/cfm/part1and2.cfm?ActType= default

Wang L.C., Lee W.J., Lee W.S., Chang-Chien G.P. and P.J. Tsai (2003). Effect of chlorine content in feeding wastes of incineration on the emission of polychlorinated dibenzo-p-dioxins/dibenzofurans, Sci. Total Environ. 302 (2003) 185–198.

Wang W-C, Lin W-H, Kuo C-P and J-Y. Wu (2012). The relation between dioxin concentration from exhaust gas of diesel engine and chlorine content. J Anal Appl Pyrolysis 2012; 94:10–6.

Wevers M., De Fre R. and Rymen T. (1992). Dioxins and Dibenzofurans in Tunnel Air. Organohalogen Compd. 9: 321–324. 2th International Symposium on Chlorinated Dioxins and Related Compounds, Tampere, Finland, 24–28 August 1992. Helsinki, Finnish Institute of Occupational Health, 1992 (Organohalogen Compounds, Vol. 9)

Xuan Z, Bi C, Li J, Nie J, Chen Z. (2017). Source contributions to total concentrations and carcinogenic potencies of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs) in ambient air: a case study in Suzhou City, China. Environ Sci Pollut Res Int. 2017 Oct; 24(30):23966-23976. doi: 10.1007/s11356-017-0050-y. Epub 2017 Sep 6. PMID: 28879468.

Appendix

- **Appendix 1**, Ctgb (2020). Technical information provided by NL CA with regards to national legislation.

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- **Appendix 2**, Position paper applicant submitted after BPC (2020). Use of products of the CMIT/MIT solvent-based biocidal product family for preservation of fuel for automotive use – Addressing concerns related to potential formation of dioxins during combustion.

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- **Appendix 3**, Position paper applicant submitted after BPC (2021). Comments to the response to the EU Commission mandate requesting ECHA opinions under Article 75(1)(g) of the BPR "Estimation of the emissions of dioxins from the use of the "CMIT-MIT SOLVENT BASED" biocidal product family (BPF) as preservative in crude oil and middle distillate fuel"

Applicant's comment.pdf

- **Appendix 4,** DE CA comment on Art. 75(1)g) request for C(M)IT_MIT SOLVENT BASED

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