Czech University of Life Sciences
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Vehicle Interior Air Quality: Review of Current Research and Legislation Status

Bachelor Thesis

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Vehicle Interior Air Quality: Review of Current Research and Legislation Status

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Objectives of thesis

The aim of my thesis is to summarize the key findings of research and studies carried out regarding the vehicle interior air quality (VIAQ) and provide an overview of the latest legislation.
This objective is motivated by the current state of knowledge in the field:

Numerous studies have found that the vehicle interior is a mixture of man-made materials that emit a vast number of chemical substances with volatile organic compounds (VOCs) being one of the major air pollutants in vehicle cabins. VOCs and other chemical compounds emitted by materials that comprise the car interior can reach very high concentrations which can have adverse effects on human health. Moreover, it has been found that prolonged exposure can lead to increased asthma and even cancer risks. The confined space of a vehicle cabin means that the combined concentrations of VOCs and other chemicals can be as much as three times higher than in other indoor environments. At the same time, there is little known about the combined effects of these chemical substances on human health as the mixture of materials and their quantity changes with every car model as well as with time, outdoor and indoor temperature, ventilation conditions and other aspects.

Given these facts regarding the noxious environment of a vehicle interior, it is quite surprising and disconcerting that very little has been done so far globally in terms of legislation governing the issue. Only a handful of countries (which are all located in Asia) have passed legislation with limits for VOCs and a few other chemical compounds commonly found in a vehicle interior. Europe, as well as the US, have so far left this area without state-imposed regulation and rely on car manufacturers' own standards regarding VIAQ.

Methodology

The thesis will be conducted using mainly the method of literature review which on some issues may be supplemented by personal written or oral interviews. I plan to research scientific databases available through the internet as well as print publications regarding the issue of VIAQ and the related aspects of air pollution, VOCs and their effects on human health. There are ample sources available. As the situation regarding the current state of legislation is a little vague and it is difficult to assess what the latest status is using only literature review, I plan to confirm my literature findings with written questioning of appropriate authorities. In some cases, it may be necessary to conduct an oral interview to reach a clear and full conclusion regarding the current legal status.

I plan to use a minimum of 50 literature sources for my thesis.

The proposed extent of the thesis

35

Keywords

volatile organic compounds, vehicle interior air quality, legislation, air pollution, health effects

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Recommended information sources

Brodzik K, Faber J, Łomankiewicz D, Gołda-Kopek A. In-vehicle VOCs composition of unconditioned, newly produced cars. Journal of Environmental Sciences. 2014 May 1;26(5):1052-61.

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Xu, Bin, Xiaokai Chen, and Jianyin Xiong. Air quality inside motor vehicles' cabins: A review. Indoor and Built Environment 27, no. 4 (2018): 452-465

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Čestné prohlášení

Prohlašuji, že jsem bakalářskou práci na téma: **Vehicle Interior Air Quality: Review of Current Research and Legislation Status** vypracovala samostatně a citovala jsem všechny informační zdroje, které jsem v práci použila a které jsem rovněž uvedla na konci práce v seznamu použitých informačních zdrojů.

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Svým podpisem rovněž prohlašuji, že elektronická verze práce je totožná s verzi tištěnou a že s údaji uvedenými v práci bylo nakládáno v souvislosti s GDPR.

V Praze, dne 1. 6. 2020

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Abstract

The car interior is a very important micro-environment that can have a serious impact on human health. It is filled with many man-made materials that all emit volatile organic compounds (VOC) at varying degrees and, considering the small volume of space in a car cabin, concentrations of VOCs can reach levels that exceed recommendations for safe exposure in interior environments many times over. Most of the organic compounds are harmful to human health and many of them are carcinogenic. Even though globally the average time spent in cars keeps increasing, very little attention is paid to vehicle interior air quality (VIAQ) by governments, legislators and consumers alike.

This paper reviewed the latest research concerning VIAQ with a focus on VOCs, their main sources and factors affecting their concentration in car interiors, as well as international legislation in regards to VIAQ. Legislation regarding VIAQ is virtually non-existent in most of the world, with China, South Korea and Japan being the only pioneers of VOC regulation aimed at car manufacturers.

Based on the research it can be concluded that there is quite a bit of room for improvement for car manufacturers who can alleviate the VIAQ situation by applying better materials and installing cabin filters with active VOC elimination. Primarily, however, car manufacturers need to be testing finished cars for VOC emissions and information about whole cabin total VOC concentrations should become available to consumers as part of basic information about vehicles they receive.

Increased consumer awareness and new legislation drafted to address this issue would ensure a safer environment in all cars, regardless of their manufacturer.

Keywords: volatile organic compounds, vehicle interior air quality, legislation, air pollution, health effects

Abstrakt

Interiér automobilu je velice důležitým mikro-prostředím, které může mít významný dopad na lidské zdraví. Je plný umělých materiálů, které do svého okolí uvolňují těkavé organické sloučeniny (Volatile Organic Compounds – VOC) v různém množství a vzhledem k malému objemu vnitřního prostředí automobilu, tak mohou koncentrace VOC dosahovat hodnot, které mnohonásobně překračují doporučené bezpečné hladiny pro vnitřní prostředí. Většina těchto organických sloučenin je lidskému zdraví škodlivá a mnoho z nich je karcinogenních. Přestože množství času stráveného v autech celosvětově roste, věnují vlády, legislativa i spotřebitelé velice malou pozornost kvalitě vnitřního ovzduší v automobilech (Vehicle Interior Air Quality – VIAQ).

Tato práce shrnuje aktuální výzkum týkající se VIAQ se zaměřením na VOC, jejich hlavní zdroje a faktory ovlivňující jejich koncentraci v interiérech aut, spolu s mezinárodní legislativou v této oblasti. Ve většině zemí světa je oblast VIAQ legislativně neregulována. Výjimku tvoří Čína, Jižní Korea a Japonsko, které jsou průkopníky, co se týče legislativy zaměřené na regulaci VOC v interiérech aut.

Z provedeného výzkumu vyplývá, že výrobci automobilů mají velký prostor pro zlepšení kvality vnitřního prostředí automobilů, ať už prostřednictvím lepších materiálů nebo instalací kabinových filtrů s aktivní eliminací VOC. Primárně, by ale měli výrobci automobilů testovat hodnoty VOC v dokončených automobilech, a údaje o celkové hodnotě těkavých organických sloučenin v kabinách automobilů by se měly stát součástí základních informací, které zákazníci obdrží o každém prodávaném automobilu.

Zvýšené povědomí zákazníků o problematice VIAQ a vytvoření nové cílené legislativy jsou cestou, která může zajistit bezpečnější vnitřní prostředí ve všech automobilech bez ohledu na jejich výrobce.

Key words: těkavé organické látky, kvalita vnitřního ovzduší v automobilech, legislativa, znečištění ovzduší, zdraví, dopady

Contents

1 Introduction

Vehicles have become an indispensable part of our lives and most people use some form of transportation every day. Car interior represents a very specific type of an indoor environment whose importance is being recognized as possibly having a significant effect on human health, which correlates with the amount of time people spend in cars. The time spent inside a vehicle varies individually depending on the part of the world and the urban or rural setting in which people live. However studies have shown a generally increasing trend in this area [Ketchum, 2019].

A recent German study on mobility reported that people spend on average 45 minutes every day inside a vehicle [Zulauf et al., 2019], while for US commuters it was 90 minutes, according to a 1997 study [Chien, 2007] ex. USEPA (1997) and Kim (2016) ex. Parry (2007) [Kim et al., 2016] reported that people can spend up to 4 hours a day in a car. Other studies have reported that people can spend from 5.5% to 16.7% of their time inside vehicles [Yang et al., 2017, Bakhtiari et al., 2018].

Car interior is regarded a specific indoor micro-environment as it is a confined space filled with many man-made materials, each composed of many chemical compounds [Liang et al., 2019]. Also, unlike in other indoor environments, the ratio of the quantity of materials to the volume of the interior space is much higher in a vehicle than in a typical building [Yoshida and Matsunaga, 2006, Faber et al., 2012]. Given their nature, each of the materials placed in the car cabin emits several types of different chemical compounds into the vehicle interior with volatile organic compounds (VOC) being the major group of them.

VOCs in general are chemical compounds that, even in relatively low concentrations, can have adverse effects on human health ranging from mild such as dizziness, headaches and nausea to more serious ones such as asthma or cancer. The most common VOCs present in a car interior include benzene, toluene, ethylbenzene, xylene (BTEX) and styrene which are all volatile and flammable aromatic hydrocarbons. Benzene is known to cause leukaemia and other haematological cancers and has no safe level of exposure [Smith, 2010], toluene has neuropathological effects and excess exposure can lead to dementia [Filley et al., 2004], xylene causes nasal and throat irritation [Rajan and Malathi, 2014] and ethylbenzene and styrene have been classified by the International Agency for Research on Cancer (IARC) as possibly and probably carcinogenic to humans. It has been established through studies that professional drivers are more prone to cancer than the general public [Rahman and Kim, 2012].

The BTEX chemicals are commonly present in outdoor air, however their concentrations in indoor air are much higher [Bolden et al., 2015] regardless of the specific environment [Guo et al., 2003] and can be more than twice as high [Hazrati et al., 2016]. The car is no exception [Brown and Cheng, 2000]. VOCs however are not the only pollutants present in the car interior and co-exposure to other air pollutants should not be neglected as passengers are commonly exposed also to particulate matter (PM), sulphur dioxides (SO_2) and nitrogen oxides (NOx) [Hadei et al., 2019].

Another negative effect of VOC emissions in the car interior is a phenomenon called "fogging." Even though it does not have a direct effect on human health it can have serious implications for car safety. Semi-volatile organic compounds with boiling points over 240 - 260 °C, mainly phthalates, condense on the windshield and together with soot and dust particles form a film that compromises visibility, especially under unfavourable weather conditions (direct sunlight, headlights in opposite direction etc.) [Bauhof and Wensing, 2008] [Brodzik et al., 2016].The oily film and the signature ”new car smell” were the first tell-tale signs of chemicals evaporating from car interior components [Shea, 1971] at the same time as the attention of scientists turned to the possibility that plastic materials were not as stable as they appeared [Shea, 1972].

The known negative effects of VOCs on human health are of concern to public health authorities and legislative norms have been put in place in many countries [Seifert, 1992, Holcomb and Seabrook, 1995] including the Czech Republic [Drahonovská and Gajdoš, 1997, World Health Organisation, 2010] to ensure that the amounts of VOCs in interior as well as exterior environments don’t exceed limits deemed safe.

However there are currently no such norms in place for the car interior environment in the Czech Republic and only a few countries worldwide have so far implemented regulations that require local car manufacturers and foreign car importers to meet certain vehicle interior air quality (VIAQ) criteria.

The world leader in these efforts is China with the most stringent VIAQ legislation and requirements regarding testing of whole vehicles and their parts for harmful and odor producing VOCs [MARKES Int., 2017]. Other countries with VIAQ legislation in place are South Korea, Japan and Russia. VIAQ in other markets is left up to self regulation by car manufacturers or their industry associations [UL, 2020].

The focus of this work is to provide an overview of air quality in the interior of passenger vehicles commercially available to common users with emphasis on VOCs presence in newly produced cars. It is key to establish the main sources of VOC pollution in the interior, the possible effects of car type and age on total VOC (TVOC) concentrations and whether there are other influential factors such as operating conditions, mode of ventilation, ambient temperature and others.

In conjunction with providing a comprehensive summary of research findings in the VIAQ area, this work will also provide an overview of legislation and norms relating to the VIAQ with a focus on the Czech Republic, the EU and selected Asian countries.

In the last three decades the VIAQ has been of growing concern to researchers who have been pointing at the elevated concentrations of air pollutants, and mainly VOCs, in the car interior, their sources and the multiple adverse effects they can have on human health. Studies have found that concentrations of VOCs in the car interior can be five to ten times higher than in ambient air and up to three times higher than in indoor air [Faber et al., 2013b] ex. [Weisel, 2005], [Jo and Park, 1999], [Xu et al., 2016] which is significantly higher than recommended for indoor environment. Other studies have shown that the exposure to aromatic hydrocarbons in cars is 2-3 times higher than in other means of transportation [Geiss et al., 2009] ex Air Quality Sciences Inc.,2006 [Dor et al., 1995].

Elevated VOC levels in car interiors can be attributed to many sources, exterior as well as interior, such as self-pollution by exhaust leaks, the intake of polluted ambient air by the A/C system, tobacco smoke, air fresheners [Fedoruk and Kerger, 2003] and others. However studies have confirmed many times over that off-gassing of new materials used in the car interior is by far the most significant source of VOC pollution in cars [Faber et al., 2013a, Yoshida and Matsunaga, 2006, Kim et al., 2016]. It has been proven that VOC concentrations increase with temperature and relative humidity [Faber et al., 2012] and decrease with car age and total mileage [You et al., 2007], with the car age being the strongest factor [Zhang et al., 2008], followed by interior temperature and driving mode [Chen et al., 2014].

And it is this off-gassing of materials in the car interior that gives new cars the very distinct "new car smell" - while considered the ultimate reward for purchasing a new vehicle by customers in the Western hemisphere, it is the number one problem for Asian customers [Krishna, 2013, J.D.POWER, 2020]. This distinct odor is however a toxic cocktail of chemicals which contains several hundred chemical compounds with VOCs representing majority of them [Agency, 2017].

During tests performed on new cars Brodzik et al. [2014] identified over 250 organic compounds in air samples tested, with aliphatic hydrocarbons representing more than 60% of them. Yoshida and Matsunaga [2006] identified at least 162 organic compounds in cars three years from production date with many aliphatic hydrocarbons and aromatic hydrocarbons as well. Zhang et al. [2008] tested 802 cars five years old and younger and found that 82% of the cars exceeded the Chinese National Indoor Air Quality Standards regarding their concentrations of benzene, toluene, xylene and formaldehyde measured.

It had been identified that the main source of VOCs in the car cabin are carpets, upholstery, leather and fabric trim on seats, hard plastics used on dashboards and steering wheels as well as sealants, glues and adhesives used to finish the interior [Brodzik et al., 2016, Fedoruk and Kerger, 2003] ex USEPA, 1976a,b; Akland and Ott,1987. Some of the off-gassing may also be due to solvents applied to clean the interior of paints and glues used earlier in the production process.

It may be sometimes difficult to determine the exact source of a certain type of a compound as it is often emitted by different materials at the same time [Faber and Brodzik, 2017]. Their amount in the air and the speed at which they are released into the car interior also changes with time lapsed since the production of the vehicle, with humidity, variations in temperature and is also influenced by interactions with other compounds present in the interior [Brodzik et al., 2014, Yoshida and Matsunaga, 2006, Brodzik et al., 2016].

Even though it is known that material off-gassing is the primary source of VOCs in car cabin, it remains a challenge to find the best way to control the TVOC for the whole vehicle as many factors can influence the outcome such as material adsorption [Huang et al., 2016], alterations in the color of components [Brodzik et al., 2014] or the simple size of the vehicle cabin [Chien, 2007].

As China pushes for cleaner and odor-free cars with its stringent VIAQ legislation and standardised testing requirements, there will be a greater need among car manufacturers as well as their suppliers to better understand and control VOC emissions of materials they use in car components [MARKES Int., 2019].

2 Research Goals

The goal of this paper is to evaluate the quality of interior air in passenger vehicles intended for individual use that are commercially sold and commonly available to customers on their respective domestic markets, and to asses the effects the quality of the car environment can have on the health of its passengers.

Information about the quality of the vehicle air and the presence of possible pollutants will be based on the research and analysis of at least 50 sources dedicated to the subject of VIAQ and carried out by researchers in different countries over the course of the last thirty years.

The aim of the research is to assess and name the main pollutants or groups of air pollutants present in the vehicle interior with a focus on volatile organic compounds and the risks they pose for human health. This paper will also determine the main source or sources of these pollutants, the factors that affect their concentrations in the vehicle air and methods used to detect and measure their presence.

Since automobile production and operation is a subject of regulation by many national and international laws and legal norms, this paper will provide a summary of the status of legislation concerning VIAQ on the international level as well as in the Czech Republic.

Drawing on the existing research this paper will try to suggest possible ways to amend and improve the VIAQ in regards to car manufacturers as well as car users. It will also provide an outlook of legislative changes that might improve the position of customers in regards to the quality of the interior vehicle environment.

3 Methodology

Findings presented in this research paper are based on the study of scientific literature related to the issue of vehicle interior air quality, air pollutants, health hazards related to air pollution, methods to measure VIAQ and legislation and automotive industry norms related to air quality inside vehicles.

Results presented in this paper in regards to the presence and sources of VOCs in the car interior are based on the analysis of a large number of practical and experimental studies carried out by various researchers in different countries and published in the time period of the past thirty years. All of the studies were peer-reviewed and published by respectable research paper databases which were searched using the Google Scholar search engine.

As the issue of VIAQ is becoming a great concern for automobile producers around the world, and mainly to those producing in and exporting to South-East Asia, there are a number of companies based in Western Europe and the USA that focus on VIAQ compliance with Asian market requirements and produce their own papers focused on the VIAQ issues. These papers, intended to educate car manufacturers or to be presented at industry-focused conferences, are a good source of the latest legislative norms and legal requirements in the main automotive markets as well as testing methods available, and will be used if needed to supplement information in this paper.

Research papers and scientific literature outside of the field of VIAQ research will be used to introduce, describe or explain facts, methods and issues concerning various other topics mentioned in the paper unrelated to the VIAQ.

Web sites and databases of a number of international organizations and automobile industry associations will be researched to find the latest information regarding air quality, health hazards and concerns, legislative norms, legal requirements and progress made on the issue of improving air quality in commercially sold vehicles.

As the issue of VIAQ is of pressing concern to automobile producers selling to South-East Asia, blog posts by industry insiders were a useful tool to confirm information obtained from scientific literature and industry association websites in regards to the latest developments concerning VIAQ legal requirements as well as to explain the strong push by Asian countries to regulate the VOC concentrations inside vehicles.

4 Results

4.1 Indoor Air Pollutants

4.1.1 Classification of Air Pollutants

Indoor and outdoor air pollutants can be classified in several ways depending on their nature, physical state, origin, particle size and other factors. Based on their origin they can be classified as biological particles (bacteria, moulds, viruses, pollen, spores etc.), non-biological particles (smoke, dust, heavy metals, radioactive isotopes, etc.) and gases (such as evaporation from adhesives, paints, cleaning products, petroleum products etc.)[Dwevedi, 2018]. Based on their physical state they can be classified as solids, liquids, gases or as disperse systems; based on their origin as organic and inorganic and so forth [Faber and Brodzik, 2017] ex [Levin, 2003].

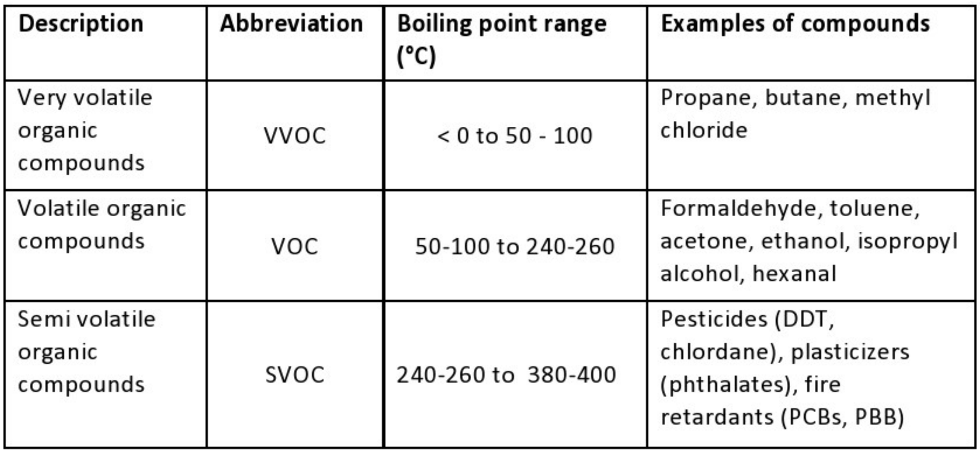
Volatile organic compounds (VOC) are low-molecular-weight organic carbon-based compounds [Gupta, 2016] whose composition makes it possible for them to evaporate under normal indoor atmospheric conditions of temperature and pressure (standard atmospheric pressure is 101.3 kPa). A big part of the VOCs are hydrocarbons which are divided into four important classes - alkanes, alkenes, aromatics and oxygenates. While the firsts three classes are generally produced during the production of petroleum-based products, oxygenates are formed in the atmosphere [Vallero, 2014a].

The volatility of a compound is higher the lower its boiling point temperature and so the volatility of organic compounds is sometimes classified by their boiling points or by their vapour pressure which is the tendency of a substance to vaporise. Substances with higher vapour pressure will vaporise more readily at a given temperature than substances with lower vapour pressure [Agency, 2017].

VOC's present in the ambient air are almost always in the gas phase since their vapour pressure in the environment is usually greater than 0.01 kPa. For VOCs then the air pollution concentration in air is almost equal to their vapour phase concentrations [Vallero, 2014b].

VOCs can be also categorised by the ease with which they will be emitted. The World Health Organization (WHO) categorises indoor organic pollutants as Very volatile organic compounds (VVOCs), Volatile organic compounds (VOCs) and Semi-volatile organic compounds (SVOCs)[Agency, 2017].

According to this classification the VVOCs, as listed in Table no.1, have a boiling point between 50 °C and 100 °C, VOCs start boiling at temperatures between 50 °C and 100 °C and end boiling between 240 °C and 260 °C, SVOCs begin boiling between 240°C and 260°C and end boiling at 380 °C and 400 °C and particulate organic matter (PM) sublimates above 400 °C [Hodgson et al., 1994].



Description	Abbreviation	Boiling point range (°C)	Examples of compounds
Very volatile organic compounds	VVOC	< 0 to 50 - 100	Propane, butane, methyl chloride
Volatile organic compounds	VOC	50-100 to 240-260	Formaldehyde, toluene, acetone, ethanol, isopropyl alcohol, hexanal
Semi volatile organic compounds	SVOC	240-260 to 380-400	Pesticides (DDT, chlordane), plasticizers (phthalates), fire retardants (PCBs, PBB)

Table no. 1 – Classification of Organic Pollutants

4.1.2 Impact of volatile organic compounds on human health

Volatile organic compounds present in air can have a range of adverse effects on human health which are linked to the amount of exposure in terms of time and concentration of the pollutants. Their concentrations also influence the level of comfort or discomfort experienced by passengers inside the vehicle cabin even if they are not always detectable by smell [MARKES Int., 2019].

As it was stated earlier there are hundreds of VOCs that can be present inside a car cabin and the most common ones are benzene, toluene, ethylbenzene, xylene (BTEX) together with styrene, formaldehyde and different phthalates. Studies strongly suggest that all four chemicals in the BTEX group have endocrine-disrupting properties at exposure levels which are below safe concentration levels issued by the U.S. Environmental Protection Agency [Bolden et al., 2015]. The BTEX are also known precursors to other air pollutants such as tropospheric ozone, particulate matter, polycyclic aromatic hydrocarbons and ultra-fine particles which all have been linked to countless negative health effects, many of which are also endocrine related [Bolden et al., 2015].

Benzene

Benzene is used as the primary material for the production of plastics, synthetic fibers, detergents, dyes, resins, drugs and pesticides [Smith, 2010]. Other significant sources are tobacco smoke, petrochemical industries in general, petrochemical products (mainly gasoline) and car transportation (combustion processes) [WHO, 1996].

According to the World Health Organization (WHO) the most adverse health effects of exposure to benzene are haemotoxicity, genotoxicity and carcinogenicity [WHO, 1996].

It has been classified by the International Agency for Research on Cancer (IARC) as carcinogenic with direct links to leukaemia, multiple myeloma, non-Hodgkin lymphoma and other haematological cancers [Cogliano et al., 2011]. It is also strongly associated with childhood leukaemia. It affects the blood forming system at low levels and there is no evidence of a safe threshold [Smith, 2010].

Toluene

Toluene (methylbenzene) can be found in paints, solvents, polishes, adhesives, metal cleaners and vinyl flooring. It is a well-studied neurotoxin with acute and chronic effects on the central nervous system (CNS).

It can cause congenital anomalies and developmental decrements and there is research to suggest it is linked to spontaneous abortions. It can also cause liver and kidney damage and chronic abuse is linked to dementia. Some of the symptoms of toluene exposure include headache, eye and nose irritation, weakness, confusion, dizziness, anxiety and muscle fatigue [for Europe, 2000] [Filley et al., 2004].

Ethylbenzene

Ethylbenzene, just like benzene, toluene and xylene, is the major component of gasoline and fuel oil [Gupta, 2018]. According to the IARC Monographs Classification of Agents, it is classified as possibly carcinogenic [IARC, 2020].

Just like other aromatic hydrocarbons, ethylbenzene is well absorbed by the respiratory and the GI track. Therefore it causes difficulty breathing, effects the upper respiratory track and the lungs, can be harmful to the kidneys, the liver and endocrine system. Effects of exposure include dizziness and even hearing loss. At very high levels, the exposure can be fatal [Epstein, 2017].

Xylene

Xylene is an aromatic hydrocarbon widely used as an industrial solvent in rubber, printing and leather industries. It can be found in paints, varnishes, cleaning solutions as well as gasoline and airplane fuel [Rajan and Malathi, 2014].

Inhalation of xylene irritates the upper respiratory tract and in high amounts can lead to impaired lung function. It affects the nervous system and causes headaches, nausea, vomiting, dizziness, fatigue confusion and can worsen short-term memory. High exposure has been linked to liver and kidney damage and, in animal studies, xylene proved to be toxic to developing fetuses [Epstein, 2017].

Styrene

Styrene is a highly volatile organic compound which in its pure form is a colorless liquid with a sweet smell. Manufactured styrene usually contains aldehydes which give it a sharp, unpleasant odor. The odor detection threshold is at 70 μg/kg however it is usually recognized by its odor at levels 3 to 4 times higher. The recommended limit for exposure before neuro-behavioural effects begin taking place is significantly lower, the WHO recommends 0.26 mg/m^3 [WHO, 1996].

Even though small amounts of styrene can be produced naturally by plants, bacteria, and fungi, the majority is manufactured and used in the production of plastics and rubber. It can be found in a wide range of products such as packaging materials, electrical insulation, plastic pipes, automobile parts and in the backing layer of carpets. It is also a result of burning and is present in cigarette smoke and automobile exhaust [ATSDR, 2020b].

According to the WHO's updated and revised air quality guidelines for Europe (1995), styrene is considered carcinogenic and genotoxic with neurological and developmental effects [WHO, 1996].

Formaldehyde

The main source of formaldehyde vapors are industrial and automobile combustion processes including heating, cooking and smoking. Among the major sources in the indoor environment are glues, paints, adhesives, varnishes, resins, insulating materials, textiles, cleaners and electronic equipment.

Exposure to formaldehyde causes respiratory tract inflammation and eye and skin irritation. At higher levels is causes skin rashes, shortness of breath, wheezing and changes in lung function [ATSDR, 2020a].

Formaldehyde has been classified as a Class 1 carcinogen and repeated and prolonged exposure has been associated with increased risk of childhood asthma, upper respiratory tract inflammation, nasopharyngeal cancer, and possibly leukaemia [World Health Organisation, 2010].

Phthalates

Phthalates are predominantly used as plasticizers in polymers to make plastic products flexible and durable. Their second application is as solvents in other products.

Phthalates can be classified based on their molecular weight:

1) **low-molecular-weight phthalates** are ester side-chain lengths with one to four carbons such as DMP, DEP, DBP, and DIBP.

They are used in a variety of personal-hygiene and cosmetic products, for example as scent stabilisers in fragrances and in nail polishes to minimize chipping.

2) **high-molecular-weight phthalates** are ester side-chain lengths with five or more carbons such as DEHP, DOP, and DINP.

They are used, for example, in vinyl floor coverings, plastic tubing, plastic containers, food packaging and processing materials, vinyl toys, medical products and building products.

Out of these two groups the most used phthalate is DEHP, which is used as a plasticizer for polyvinyl chloride (PVC) polymers which account for 90% of world production of DEHP. Products may contain from 1% to 40% of DEHP by weight [for Research on Cancer et al., 2012]. World consumption of phthalates in the early 1990s was estimated to be 3.25 million tonnes, of which DEHP used in plastics production accounted for approximately 2.1 million tonnes.

Phthalates used as plasticizers in PVC plastics are not chemically bound to the polymers and they can easily leach, migrate or evaporate, especially when phthalate-containing products are exposed to high temperatures [Council et al., 2009].

Off-gassing from consumer products containing phthalates can result in human exposure either through direct contact or indirectly through leaching into other products and can take place via ingestion, inhalation and skin contact. Some phthalates are known reproductive and developmental toxicants in animals and suspected endocrine disruptors in humans [Heudorf et al., 2007].

It had been established through both animal and human studies that exposure can affect humans throughout their life span including in-utero development as phthalates can cross the placenta, have been measured in amniotic fluid in human studies and are present in breast milk [Council et al., 2009].

Naphthalene

Naphthalene is a solid volatile substance, an aromatic hydrocarbon, which is derived from coal tar during the process of distillation. In terms of chemical properties it is very similar to benzene and is most commonly used as a household insecticide, for example in moth balls. It is also a product of combustion processes so it can be found in tobacco smoke, vehicle exhaust fumes, forest fires smoke etc [Irwin et al., 1997].

It has a very high toxic potential and effects of acute exposure include headaches, dizziness, nausea, vomiting and abdominal pain. Chronic exposure can cause liver, kidney, retinal and neurological damage as well as developmental and neurological damage to fetuses. Another effect of naphthalene exposure is hemolytic anaemia [Gupta, 2016, Jo and Lee, 2011, Mazzaferro and Ford, 2012, National Pesticide Information Center, 2020].

4.1.3 Sick Building Syndrome (SBS) - parallels with ”New Car Smell” and TVOC levels in new cars

Similar to their nauseating effect in cars, volatile organic compounds have also been identified as one of the main pollutant categories behind the ”Sick Building Syndrome” or SBS; the other group responsible being aerosols. It is no accident that exposure to elevated levels of VOCs in buildings, which is what SBS is caused by, are responsible for the same type of discomfort suffered by some passengers in new cars [Redlich et al., 1997].

SBS symptoms have been defined to include mild forms such as eye, nose and throat irritation, headaches, fatigue and difficulty concentrating, to more severe forms such as asthma, allergies or hypersensitivity pneumonitis [Hodgson et al., 1994]. The World Health Organization (WHO) recognised SBS as a medical condition in 1982 and according to Jansz (2017) ex. Hedge and Ericson (1996) [Jansz, 2017] the syndrome seems to be linked with occupancy of certain workplaces.

A study carried out in older cars and public buses in Poland found that formaldehyde and acrolein, compounds responsible for the SBS, were present in every vehicle and bus tested however none of the values exceeded recommended exposure levels [Dudzińska, 2011]. It is important to mention though that many of the SBS symptoms have been observed at exposure levels two to three levels of magnitude below permitted levels and threshold limit values (TLV).

Seifert and Ullrich [1987] for example recommend a level of 0.25 mg/m^3 while Mølhave et al. [1986] suggests 0.3 mg/m^3 of TVOC as an acceptable level, concluding that higher exposure can cause problems [Hodgson et al., 1994]. It had been observed that exposure to a mixture of 22 VOCs commonly found in indoor spaces caused measurable symptoms of discomfort and decrease in attention at levels of 5 mg/m^3 as well as 25 mg/m^3 [Mølhave et al., 1986].

While there are studies that have found BTEX levels in office buildings in the range of 0.005–0.047 mg/m^3 to be considerably higher than the corresponding outdoor levels [Horemans et al., 2008], TVOC levels in new cars mentioned in this work range between 14 to 64 mg/m^3 [Yoshida and Matsunaga, 2006, Brown and Cheng, 2000].

4.2 Sources of VOCs in car interior

Several sources of volatile organic compounds in the car interior have been recognised:

1) off-gassing from materials used to equip the car interior

2) self pollution from the car's own exhaust system; intake of emissions present in the ambient environment

3) impact of users personal habits and preferences - tobacco smoking, air fresheners, spills of various materials, introduction of other VOC emitting materials etc.

The presence of volatile organic compounds in the car interior is mostly the result of off-gassing from materials used to equip the car cabin [Yoshida and Matsunaga, 2006, Yang et al., 2020, Janicka et al., 2010], however the effect of other sources cannot be neglected [Chien, 2007]. Depending on a particular driving scenario the combined effect of all of the above elements can result in VOC concentrations elevated way above levels deemed safe by health norms for interior environment [Faber et al., 2013b].

4.2.1 Off-gassing from materials used to equip the car interior

The interior of a car cabin is filled with components that are made out of various materials and most are composites of different compounds. Even though self-pollution with the car's own exhaust fumes can add to the concentrations of BTEX and styrene, which are generally measured to determine the VIAQ and are also markers for gasoline fuels [Soldatos et al., 2003, Serrano-Trespalacios et al., 2004], tests carried out under static conditions and without air exchange with the ambient environment confirmed that it is the interior materials that are responsible for a large portion of TVOCs in a car [Faber et al., 2014, Chien, 2007].

Many studies established that materials used in the car interior are the biggest source of VOCs in the cabin and that their exact composition, finish and combination can significantly affect the total concentrations of VOCs [Brodzik et al., 2014, Zhang et al., 2008, Chien, 2007].

The TVOCs are the highest in newest cars and decrease with time and good ventilation [Yoshida and Matsunaga, 2006, Chien, 2007]. It has been established that VOC emissions increase significantly with temperature and that other factors affecting TVOC concentrations are humidity, driving mode and overall technical condition of the vehicle [Faber et al., 2014, Kim et al., 2016, Yang et al., 2020].

VOCs are emitted by all of the components and materials present in the vehicle cabin and the size of their exposed surface area is in direct proportion to their effect on the TVOC concentrations. Therefore since the seats, ceiling upholstery, door paneling, the dashboard and carpets make up the largest proportion of surface area, their material emissions can have the biggest effect on total VOC concentrations [Brodzik et al., 2016]. Testing has shown that materials with low emission strength can produce high VOC levels if they cover large surface areas [Yang et al., 2020].

Additionally during the production process many materials are covered with paints and sealants, affixed with glues and treated with lubricants, cleaning substances, polishes, surface treatments, deodorizers and other chemical substances that aim to improve the user's visual enjoyment and comfort level in the car, yet add to the toxic cocktail of chemicals that subsequently fill the air in the cabin [Chien, 2007, Fedoruk and Kerger, 2003, Brodzik et al., 2016].

While some of the VOCs are a product of the manufacturing process and are emitted into the environment continuously (off-gassing), other ones may occur later as the result of aging-related breakdown, change in temperature (heating/cooling), etc. [Fedoruk and Kerger, 2003].

Components in the car interior are made out of a wide range of materials, the most common ones being: polystyrene, polypropylene, polyethylene, polyamid, phthalates, acrilonitrile-butadiene-styrene (ABS) latex glue, polyurethane foam, natural and synthetic leather [Geiss et al., 2009, Brodzik et al., 2016, You et al., 2007].

Each of these materials is a source of very specific VOCs that are emitted under certain conditions. For example, at 23 °C polyethylene releases diethyl phthalate, benzaldehyde, tridecane, nonanal and decanal, and at a higher temperature of about 70 °C also 2-ethyl-1-hexanol. Polystyrene is the source of ethylbenzene and styrene regardless of ambient temperature, and polyurethane foam emits toluene, benzaldehyde, nonanal, phenol, decane, diethyl phthalate and others [Brodzik et al., 2016]. Similarly aromatic hydrocarbons are the markers of upholstery emissions, with toluene, ethylbenzene and isomers of xylene being the compounds with the highest ratio [Brodzik et al., 2014].

Most of these materials consist of volatile organic compounds which due to their low boiling points are highly volatile at temperatures that can normally occur inside a vehicle and their volatility increases with rising temperature [Geiss et al., 2009, Faber et al., 2012].

With respect to time, emissions from materials can be divided into primary and secondary. Primary emissions are those released within the first year of the material's production and are generally low-molecular-weight compounds such as additives, solvent residues and monomers. Secondary emission are those that occur after the first year after the material was introduced into the indoor environment and are generally the products of degradation, decomposition, hydrolysis, oxidation and other reactions [Kataoka et al., 2012]. The concentrations of compounds resulting from secondary emissions are usually significantly lower than from primary emissions [Faber and Brodzik, 2017] ex. Zabiegala B(2006).

Aware of the VOCs' effect on VIAQ, various studies have subsequently tried to point out what parts of the car interior are responsible for most of the VOC emissions, what specific organic compounds they emit and whether variations in the final finish of the material can influence the results. Tests carried out on the same models with different interior trim or different models with different trim by the same manufacturer established that the interior equipment affects the presence of certain VOCs and their concentrations on a noticeable level. The variations in VOC levels between different brands are also statistically greater than variations between different models of the same brand which some researchers contributed to un-tested factors such as the influence of the cabin's layout [Chien, 2007].

Similarly Brodzik et al. [2014] observed that the intra-model variability in total VOC levels in two identically equipped cars by the same manufacturer is not more than 14% indicating that a certain combination of materials and equipment can help predict TVOC levels in a car cabin. The number of organic compounds detected in a vehicle is also in positive correlation with the TVOC concentrations thus allowing the indication of elevated TVOC levels based on enhanced emissions from interior materials.

Assessing the influence of interior trim on TVOCs, Xu et al. [2016] found a positive correlation between leather trim seats and higher VOC concentrations compared to fabric trim seats . The mean concentrations of the main pollutants measured, such as benzene, toluene, ethylbenezene, styrene, xylene, formaldehyde, acetaldehyde, acroleine and acetone, were higher in cars with leather trim seats in all instances with toluene concentrations showing a 39% increase. The connection between leather seats and increased TVOC was the same in all vehicles regardless of age and ventilation mode used. Bakhtiari et al. [2018] confirmed Xu's findings when he also measured higher concentrations of formaldehyde and acetaldehyde in cars equipped with leather seats.

The automotive industry is aware of the problem with increased VOC emissions from leather trim components and the International Union of Leather Technologists and Chemists Societies (IULTCS) at its 35th congress in Dresden, Germany in 2019 admitted to having an issue with acetaldehyde emissions in particular [Rabe et al., 2019]. Being pressured by the latest Chinese VIAQ Standards, which are the toughest in the world, the automotive leather industry was able to lower the BTEX and styrene emissions below the imposed limits. Acetaldehyde emissions however remain in most cases higher than the required 0,2 mg/m^3.

When testing five brand-new cars Faber et al. [2013a] found the lowest VOC concentrations of identified aliphatic compounds, aromatic compounds and cycloalkanes in a car equipped with fabric-covered seats and a rubber covered steering wheel (0.313 mg/m^3, 0.207 mg/m^3 and 0.151 mg/m^3 respectively) as opposed to alcantara-covered seats and a black leather wheel which showed the highest concentrations (4.026 mg/m^3, 0.783 mg/m^3 and 1.341 mg/m^3 respectively).

Not only the material used but also different manufacturing processes and the use of different solvents, paints, glues and sealants affects the concentrations of VOCs in the cabin. While cars by one manufacturer measured higher TVOCs for cars with leather trim compared to fabric trim seats, cars by another manufacturer showed higher VOC levels for fabric trim seats than leather seats [Chien, 2007].

Even though automobile manufacturing is a highly automated process with computers controlling most of the steps of production, glitches in the process can lead to irregularities in the total VOC levels and two identical cars from the same manufacturer can show a difference in TVOC level of more than 200% [Brodzik et al., 2014].

Another factor that varies among car manufacturers and even among models of the same manufacturer is the quality of materials used to make the upholstery of the seats, plastics of the dashboard, headlining, carpets and other interior components. Studies have found that there can even be a correlation between the quality of the materials used and the sale price of the vehicle even though not very strong.

Zhang et al. [2008] tested 802 new cars in Beijing, China and found that low-end cars had higher concentrations of formaldehyde, benzene, toluene and xylene which were the target organic compounds of the study suggesting that high-emitting materials are more often used in low-end cars. Those cars were generally equipped with synthetic fabric seats, a synthetic rubber steering wheel and low grade carpet. The lesser quality of the materials used in the low-end cars was also indicated by their strong odors which their owners complained of. Surprisingly the difference between mid-range vehicles and the high-end vehicles TVOC was not very significant and the concentration of the target compounds were higher in the more expensive high-end vehicles than in the mid-range group [Zhang et al., 2008].

A large percentage of the tested vehicles also failed the Chinese National Indoor Air Quality Standard with 82% of vehicles exceeding the toluene levels, 75% exceeding the benzene levels, 25% exceeding the xylene levels and 24% of the vehicles exceeding the formaldehyde levels [Zhang et al., 2008].

A similar test looking at the correlation between price and material quality carried out by Janicka et al. [2010] compared high-end and low-end vehicles for total VOC concentrations. In contrast to the previous study, however, it showed that a higher price tag does not always guarantee better VIAQ.

A high-end vehicle, low-end vehicle and a cargo van adjusted for passenger transportation were tested for total VOC and BTX concentrations with surprising results. The high-end vehicle showed the highest TVOC at 3.9 mg/m^3, while the low-end vehicle came in second with 2.5 mg/m^3. The rebuilt transportation van had the best results with TVOCs at 2.29 mg/m^3, more than a 40% decrease compared to the most expensive vehicle of the three [Janicka et al., 2010].

Even though the study did not mention this possibility, it can be only assumed that the bigger interior space of the transportation vehicle influenced the outcome of the tests by providing a better space volume to TVOC ratio as some other studies suggested may have a positive effect [Chien, 2007]. Similarly, a study that tested certain VOC concentrations in older cars and public buses in Poland, found overall levels to be lower in the newer buses (approx. 2 years of age) than in the much older cars (4 to 12 years of age), even though new cars generally show TVOC levels exceeding recommended exposure limits many times over [Dudzińska, 2011].

Emitted VOCs are so material specific that even alterations in color of the same material affect the types of organic compounds measured and their levels [Faber et al., 2014, Brodzik et al., 2014]. When testing cars with synthetic leather and synthetic fabric, Brodzik et al. [2014] measured twice as high mean TVOCs for cars with black and white synthetic fabric and white synthetic steering wheels than for cars with combination of red fabric and white synthetic leather seats and white synthetic steering wheel - 2.49 and 3.04 mg/m^3 for the black and white combination compared to 1.35 and 1.42 mg/m^3 for the red and white interior cars.

A difference was noted also in the number of volatile organic compounds present as vehicles with black and white synthetic fabric showed a 30% higher count of total compounds than vehicles with a predominantly white interior. The lowest TVOC was noted for cars with combination red fabric, white synthetic leather and white synthetic steering wheel indicating that materials with black color dye emitted more VOCs than any other color combination [Brodzik et al., 2014].

Faber et al. [2014] tested the difference between vehicles equipped with different materials and different color combinations of the same materials and found out that while the number of compounds detected in both groups was quite similar - 228 and 200 respectively - there were some that were specific only for a certain type of material.

For example cars that had black-gray fabric upholstery, black plastic steering wheel and a gray dashboard all showed styrene, trimethylbenzene, 2-butanone and dimethyloctane in their air samples. On the other hand cars with black suede upholstery, black leather-like steering wheel and black dashboard were the only ones with positive tests for hexane, methylcyclopentane, 2,5-dimethylpentane and 2,6-dimethylcyclopentane. Compounds such as methylcyclohexane, octane and heptane were measured in both types of interiors however their concentrations were higher in the first group of cars (with fabric upholstery).

More than half of the compounds measured by Faber et al. [2014] were found in 85% of the air samples tested and about 50% of these compounds were identified which translated to the conclusion that at least 50% of the vehicles' VOCs were emitted by the materials used in the vehicles interior.

It can be assumed that compounds that were present in both types of interiors at about the same concentrations such as benzene, ethylbenzene, toluene, ethyltoluene, naphthalene, isomers of xylene, butyl and some others were emitted by interior parts that were identical for both groups of vehicles such as carpets and headlining. The total VOCs measured in the two groups of vehicles were 2.109 mg/m^3 and 1.505 mg/m^3 respectively and such levels can be already responsible for some irritation and discomfort for passengers [Faber et al., 2014, Schupp et al., 2006].

4.2.2 Self pollution from the car's own exhaust system; Intake of emissions present in ambient environment

Self-pollution also called self-exposure or back diffusion is defined as the intrusion of the vehicle's own exhaust fumes into the passenger compartment [Abi-Esber and El-Fadel, 2013, Rahman and Kim, 2012] and can be a significant contributor to the air pollution in the car interior. It can occur by the car's own engine fumes entering the vehicle cabin directly through micro-cracks in the car bodywork instead of leaving the car engine through the exhaust pipe.

Additional pollution can be introduced into the car cabin by air exchange which takes place between the vehicle and the outside environment either via the car's ventilation system or through opened windows. This air can contain both the car's own exhaust fumes as well as pollution present in the ambient air.

Also refueling has been found to have an immediate negative effect on VOC levels inside the car cabin [Yoshida and Matsunaga, 2006, Bakhtiari et al., 2018]. One way is the entry of the VOCs and mainly BTEX compounds through doors and windows after they are opened at a refueling station, the other is the burning of the fuel which introduces pollutants through back diffusion.

Even though some studies indicated that leaks of polluted air from the car's own engine compartment into the cabin were problems associated mainly with older cars, other studies that focused specifically on this phenomena reported that intrusion of fumes takes place irrespective of the vehicle's age [Abi-Esber and El-Fadel, 2013].

Leung and Harrison [1999] in a series of tests established that pollution present in the ambient air has a strong effect on the VIAQ and that the outdoor pollution is mirrored in the air quality in the car irrespective of ventilation mode selected. Still, older cars seem to show a higher intake of emissions present in the ambient environment caused either by leaks in the car's body or by lower combustion efficiency of older vehicles [Jo and Lee, 2011, Leung and Harrison, 1999].

Back diffusion was found to be a strong factor behind increased concentrations of certain volatile organic compounds and carbonyls such as benzene, toluene, formaldehyde, acetaldehyde, butylaldehyde and propionaldehyde as they were measured strictly in correlation to full speed driving mode. The same emissions were fairly insignificant just after starting the engine. Toluene concentration levels, which can be used as the strongest marker indicating back diffusion, showed an almost 90% difference between a driving mode and a static mode [Rahman and Kim, 2012].

Also higher summer temperatures seem to affect the level of fuel fumes and back diffusion being drafted into the cabin, the same correlation exists between increased temperature and VOC emissions from materials in the car cabin (see Chapter 4.3.3.).

Jo and Park [1999] found benzene and toluene levels to be almost 80% higher during summer driving compared to winter driving on the same urban routes regardless of ventilation mode used. Since benzene and toluene are highly volatile compounds, the increased summer temperatures would lead to their higher engine running loss and so they would have a higher influence on the interior air profile.

A study that focused on in-vehicle naphthalene levels based on the type of fuel used determined that naphthalene concentrations changed significantly depending on whether the vehicle was diesel-fueled or gasoline-fueled [Jo and Lee, 2011]. Since each type of fuel has its own specific set of VOCs that act as markers, elevated levels of these organic compounds in the interior point at the vehicle's own combustion fumes being diffused back into the car cabin.

While naphthalene is predominant in diesel fuel, diesel-powered cars would register higher levels of this compound in their cabins as a result of the unburned fuel fumes entering its interior. On the other hand, gasoline-powered cars showed elevated levels of the so called monocyclic aromatic hydrocarbons (MAH) which include benzene, toluene, ethylbezene and xylene, compounds added to gasoline to increase its octane number. In contrast to that vehicles using CNG or LPG fuels show BTEX concentrations below detection limit [Bakhtiari et al., 2018]. These results show that the choice of fuel can affect not only the vehicle's emissions but also the exposure to air pollutants.

Naphthalene concentrations in gasoline-fueled cars ranged between 1.13 and 5.36 mg/m^3 with mean at 3.97 mg/m^3 while in diesel-fueled cars the range was between 3.14 and 8.58 mg/m^3 with mean at 5.53 mg/m^3. The nearly 40% difference in naphthalene's mean level being a clear marker of a back diffusion of diesel cars with their own fumes.

The same trend was recorded for gasoline-fueled cars which showed benzene levels between 6.52 and 33.6 mg/m^3 with mean at 15.1 mg/m^3 compared to the diesel-fueled cars with 2.77 to 21.9 mg/m^3 range and mean of 9.4 mg/m^3, a 60% difference between the two types of vehicles [Jo and Lee, 2011].

A correlation between the level of back diffusion and CO levels was observed

[Rahman and Kim, 2012] with CO presence having a negative effect on the rate of back diffusion. With elevated CO emissions (greater than 10 ppm) also the in-vehicle TVOC concentrations increased due to a higher degree of back diffusion.

Temperature and pressure gradients have been reported to affect pollutant infiltration factors in buildings with minimal difference between in an out pressure and inside/outside temperature resulting in minimized particle infiltration rates [Abi-Esber and El-Fadel, 2013]. In the case of a moving vehicle the difference in inside/outside pressure can reach up to 5000 Pa and with air conditioning on the temperature difference can be quite significant as well [Qi et al., 2008].

Therefore it can be expected that the difference between pressure and temperature inside and outside the car cabin can induce air flow through cracks and openings thus bringing in more pollutants from the ambient air, however such expectation have not yet been quantified [Abi-Esber and El-Fadel, 2013].

Another factor that can contribute to higher infiltration of ambient air pollutants into the car interior is the height of the vehicles. As studies comparing the VOC concentrations in buses and cars have documented, the lower the height of the vehicle, the higher the intake of pollutants into the car cabin as most of the volatile compounds tend to settle close to the ground [Jo and Lee, 2011, Jo and Park, 1999].

Studies that examined the effect of driving environment and ventilation mode on the in-vehicle concentrations of CO, NO and NO_2 found that there was a strong correlation between those two factors and the level of air pollutants introduced into the car [Chan and Chung, 2003].

In urban areas and in heavy traffic situations driving with opened windows or with air conditioning on fresh air intake brought indoor pollution concentrations to the same or very similar levels as they were in the ambient environment. For example dust concentrations inside the car can be quite severe and even exceed the outdoor concentrations by three to five times [Chan and Chung, 2003] ex. Alm et al (1999).

Driving with A/C on recirculating mode decreased the amount of outdoor pollution entering the vehicle however lead to buildup of indoor pollutants already present in the vehicle. This can be a problem with CO concentrations as well as VOCs emitted from the interior components.

Another factor that can affect the amount of back diffusion is the size of the vehicle, with smaller cars showing a smaller intake of car's own exhaust fumes. A study that compared the effect of driving mode on VOC levels in a car interior found that smaller and mid-size cars showed lower TVOC levels than a large sedan by the same manufacturer when idling [Kim et al., 2016].

Even though all three types of vehicles confirmed the trend of TVOC spike once the engine was turned on, while benzene levels for small and mid-size models grew by 100%, the large-sedan's benzene concentration grew by 400 - 600% [Kim et al., 2016]. Both types of vehicles also reached comparatively higher values (4.63 - 6.03 versus 1.43 - 2.42 ppb). Other aromatic compounds such as xylene, styrene and toluene raised over the ambient air levels in a more gradual fashion.

4.2.3 Impact of users' personal habits and preferences

Personal habits and preferences of car users represent a very unpredictable yet very significant factor that can contribute to the overall concentrations of volatile organic compounds in the car cabin. Their frequency and intensity can have a varying effect which can oscillate between negligible to very high and together with other known factors that contribute to VOC concentration in cars (interior materials emissions, back-diffusion, outdoor pollution and refueling) can further deteriorate the VIAQ.

Among personal habits that contribute to increased concentrations of volatile organic compounds in the car cabin are smoking, use of air fresheners, choice of fuel, the lack of a catalytic converter, the frequency and type of car care products used (upholstery shampoos, vinyl treatments, glass cleaners, body work waxes, polishes etc.) and ad hoc events such as spills of chemical substances in the car interior, transportation of cargo that emits high amounts of VOC etc [You et al., 2007, Leung and Harrison, 1999].

Smoking

Smoking i.e. combustion of tobacco products is the source of a complex aerosol containing more than 4400 chemicals that include high amounts of volatile organic compounds such as styrene, m,p,o-xylene, benzene, ethylbenzene and 1,3,5-trimethylbenzene. Whole cigarette smoke has been classified by IARC as carcinogenic with many of the individual chemicals being biologically active as carcinogens, teratogens, or with implications for cardiovascular disease [Polzin et al., 2007].

It has been determined that individuals exposed to second-hand smoking or environment tobacco smoking (ETS) were exposed to approximately 1 - 3 times higher concentrations of these VOCs than non-exposed individuals and that ETS contributed to more than 70% of personal VOC exposure compared to other common sources such as vehicle emissions, industrial sources, recent house renovation and chlorinated water [Zhou et al., 2011]. In another study, cars with at least one passenger smoking showed benzene exposure levels elevated by 57% and toluene exposure levels by 16% [Leung and Harrison, 1999].

Risk assessment studies show that four VOCs, more specifically acetaldehyde, 1,3-butadiene, benzene and acrylonitrile, are among the top five compounds in mainstream cigarette smoke with the highest cancer risk indices. The other being arsenic and acrolein which has the highest non-cancer risk index for respiratory effects.

Benzene, which is formed during incomplete combustion, is generated at average levels of 431 μg per cigarette (263–590 μg per cigarette), 1,3-butadiene is generated at an average of 279 μg per cigarette (157–400 μg per cigarette) and acrylonitrile is emitted at 170 μg per cigarette (99–250 μg per cigarette) [Helen et al., 2014].

Air fresheners

Another significant contributor to in-vehicle VOCs are air fresheners commonly placed in vehicle cabins by car users. They are offered to consumers as a way to "freshen the air" in the car while in fact they just serve the purpose of masking the odors in the interior perceived by passengers as unpleasant. And what is worse, they further worsen the air quality in the vehicle [Duh, 2015].

Studies that focused on the health impact of using air fresheners in indoor spaces found that air fresheners not only fail to remove any contaminants from the indoor air but contrary to the common belief, add toxic chemicals to the air which may lead to severe health problems for the occupants [Perera et al., 2013].

Air fresheners work on the principal of using a nerve-deadening agent which interferes with the sense of smell by coating the nasal passages with an oily film that subsequently masks the unpleasant odor with another odor perceived as pleasant or by completely deactivating the offending odor [Jo et al., 2008].

Air fresheners may contain as many as 133 different VOCs [Potera, 2011] with many of them being toxic or hazardous. For example toluene, benzene, ethyl benzene and xylene are known carcinogens, sensitizers, neurotoxins and possible reproductive toxins [Jo et al., 2008]. Many of them also contain phthalates which are known to cause endocrine abnormalities, birth defects and reproductive problems and their only purpose in the AF is to enhance and maintain the smell of the air freshener [Perera et al., 2013].

A study that focused on endocrine disruptors in consumer products [Dodson et al., 2012] found the highest concentrations of DEP (lower molecular weight phthalate used as a solvent) in fragrances (14,000 μg/g) and car air fresheners (8,000 μg/g).

Other scent carriers commonly detected in AF are acetone, ethanol, limonene and α and β pinene [Perera et al., 2013]. It has been found that it is not uncommon for air fresheners, as well as other household products, to contain chemical compounds that are not listed [Perera et al., 2013, Dodson et al., 2012].

Other compounds of concern are limonene and linalool, also commonly found in AFs, that are known to be unsaturated ozone-reactive VOCs [Jo et al., 2008]. Limonene and linalool belong to the group of chemicals known as terpenes and are commonly utilised in household products due to their favourable odor and solvent properties. Terpenes are of great concern because when they react with ozone they form secondary pollutants such as hydroxyle radicals, formaldehyde, nitrogen oxides, hydrogen peroxide and organic aerosol.

For example a test of car air fresheners sold in South Korea identified over 80 different chemical compounds with limonen being present in 58% of all the products and linalool in 35% of all the AF tested [Jo et al., 2008]. Other commonly found substances were toluene, acetylene, benzene, hexamethylcyclotrisiloxane, pentadecane, ethanol, ethyl benzene and xylene in decreasing order of frequency.

Car care products

Another route for VOCs and other harmful chemical compounds into a car cabin is via car care products such as shampoos, cleaners, waxes, polishes etc. DBP and benzylbutyl phthalate (BBP) were detected in a car polish/wax and a car interior cleaner, cyclosiloxanes (cyclic volatile methylsiloxanes), which are added to consumer products to enhance conditioning and spreading, were found in a conventional car interior cleaner (<100 µg/g).

Cyclosiloxanes appear to be persistent and have relatively long half-lives in humans [Dodson et al., 2012]. Alkylphenol polyethoxylates (APEOs) which are used as surfactants in consumer products, were also found in car cleaners, even though in very low concentrations (< 20 µg/g).

Car fuel choice

Due to the effects of back diffusion and car cabin leaks, the choice of a car fuel can have an effect on VOC emission in the vehicle as well. While gasoline is a major source of BTEX emissions, it is below detection limit in diesel and CNG fuels [Bakhtiari et al., 2018, Thiruvengadam et al., 2016] and at very low levels in LPG fuels [Myung et al., 2012]. On the other hand, natural gas emissions contain formaldehyde and acetaldehyde in very high concentrations [Bakhtiari et al., 2018] and diesel fuel is a major source of naphthalene emissions [Jo and Lee, 2011].

Catalytic converter

Similarly, cars equipped with a catalytic converter show lower in-cabin VOC emissions compared to cars without them even though elevated VOCs in cars without catalytic converters can be partially associated with body cracks and poor maintenance due to old age (cars over 10 years old) [Som et al., 2007]. While one study showed in-car concentrations of benzene in older cars without converters to be twice as high as in newer cars with converters [Duffy and Nelson, 1997], another study put the difference at seven-fold [Som et al., 2007].

4.3 Factors affecting total volatile organic compounds concentrations

4.3.1 Vehicle age

It is no accident that new cars come with the strongest ”new car smell” as it is these brand-new vehicles just off the production line that display the highest quantity and concentrations of volatile organic compounds measured. Numerous studies that focused on testing vehicles of different age concluded that the highest concentrations are present in cars that are within days or weeks of manufacture. They also observed that these concentrations have a tendency to decrease with time [Faber et al., 2014, Yoshida and Matsunaga, 2006, You et al., 2007], however their complete elimination is impossible [Johansson, 1999].

The time lapsed from date of manufacture plays a very significant role in the amount of total VOC concentrations as the decreasing of TVOC is an exponential process with every week contributing to the overall decline by approximately 20% [Brown and Cheng, 2000].

A study that compared three vehicles at the age of three weeks, ten weeks and sixteen weeks from manufacture found that the difference of thirteen weeks between the newest and oldest vehicle meant 30-fold difference in TVOC concentrations [Brown and Cheng, 2000]. While the newest car demonstrated total VOC concentrations at 64 mg/m^3, the ten-week-old car was at 20 mg/m^3 and the sixteen-week-old car recorded only 2.1 mg/m^3 TVOC. That is still however more than eight times higher than the exposure level of 0.25 mg/m^3 recommended by Seifert [Seifert and Ullrich, 1987].

Another study, that focused on the effect of the so called ”fogging” (caused by emissions of semi-volatile organic compounds in the car interior adhering to windshields and thus causing decreased visibility), tested new cars and the effect of time on total SVOC concentrations by artificially aging the cars. While the concentrations of selected SVOCs in the new cars ranged from 7 to 24 mg/m^3, cars "aged" by having interior temperature raised to 65 °C for eight hours every day showed after 20 days of such treatment levels that were 2.4 to 2.8 times lower (2.5 - 10 mg/m^3) than at the beginning. Further tempering brought the total concentrations after 20 more days another 2.2 - 2.5 times lower to levels of 1 - 4.5 mg/m^3 [Bauhof and Wensing, 2008].

Bauhof and Wensing [2008] also identified that different organic compounds are responsible for the new car smell as opposed to fogging. While fogging is caused mainly by semi-volatile compounds such as higher fatty acids, paraffins, phthalates, esters, phosphoric acid esters, halogenated hydrocarbons, organic silicon compounds, oxygen, nitrogen and sulfur compounds, the "new car smell" was the result of volatile organic compounds that include alkanes, carbonyl compounds, aromatic hydrocarbons, alcohols, residual monomers, ethers, esters, halogenated hydrocarbons, terpens, nitrogen and sulfur compounds.

It can be debated whether such controlled process can properly simulate the effect of real life aging which is normally affected by many more factors such as air exchange, ambient air pollution, effect of ambient weather conditions and frequency and length of driving coming into play. The study also did not stipulate whether the "aging process" was equivalent to 20 days of real life use or a different time period. Whatever the exact time equation of the test conditions, the study confirmed that there is a positive correlation between time and total concentrations of volatile compounds.

The total VOC concentrations respond to time also in a much shorter time window. Tests that focused on the change of TVOC concentrations at different temperatures over the span of 26 hours showed that regardless of the temperature, the VOC emissions always followed the same pattern of increase, peak and decrease [Yang et al., 2020].

The trend of emissions decreasing in correlation to time was recorded also by You et al. [2007] who tested three vehicles of different age (new, 1 and 5 years old) under static conditions to determine the quantity and concentrations of TVOCs . In two vehicles of the same model, the new vehicle had four times the total VOC concentrations (4.94 mg/m^3) compared to the one-year-old model (1.24 mg/m^3) which You et al. [2007] attributed to the air exchange that took place in the car since delivery. The five-year-old car, which was a different model than the other two tested vehicles, showed 40 times lower (0.132 mg/m^3) concentrations compared to the brand new automobile.

Yoshida and Matsunaga [2006] found more than 162 VOCs in the interior of a brand new car no more than two weeks from manufacture with aliphatic hydrocarbons and aromatic hydrocarbons representing 90% of the organic compounds detected. More specifically the aliphatic hydrocarbons made up 51.8% of the total concentrations at 7.294 mg/m^3 while aromatic hydrocarbons scored 42.2% at 5.947 mg/m^3. The rest of the VOCs were at almost negligible levels such as 0.4% (0.057 mg/m^3) for carbonyl compounds, 0.3% (0.044 mg/m^3) for esters, 0.2% (0.022 mg/m^3) for halocarbons, 0.1% (0.009 mg/m^3) for terpens and 5% (0.709 mg/m^3) for others. The samples were taken at interior temperature of 36 °C and relative humidity of about 77%.

The study [Yoshida and Matsunaga, 2006] also confirmed that the TVOC declined with time. While the sum of all the concentrations determined in the study was about 14 mg/m^3 on the day after delivery, a year later the TVOC was about one tenth of the original measurements. The long-term reduction of these concentrations however slowed with time and only a slight decrease was recorded after the second year of this three-year study.

The time since manufacture affects not only the concentrations but also the count of organic compounds. While the number of substances recorded in a new car topped 82 organic compounds in one study, their count dropped by 25% to 61 substances in a one-year-old car and to "only" 36 compounds in a five year-old-car which was more than a 56% decrease, compared to the brand new vehicle [You et al., 2007].

The dominant VOCs detected in vehicles by most studies included toluene, xylene, styrene, ethylbenzene, acetone and benzene derivates which are common in paints, sealants, carpets and adhesives used in the interior. For example styrene was generally found as part of styrene-butadiene rubber (SBR) glue which is used in the production of carpets [You et al., 2007, Grabbs et al., 2000, Faber et al., 2013b, Brown and Cheng, 2000].

Aliphatic compounds also dominate the list of VOC collected in new vehicles with undecane, methyldecane, methylhexane and heptane being present in significant concentrations [Faber et al., 2013a]. Aliphatic, aromatic and cyclic compounds accounted for almost 90% of all VOCs identified by Faber et al. [2013a] in a new car test with their concentrations ranging from 0.797 to 6.711 mg/m^3.

Another study found over sixty different chemicals in the interiors of four new cars with their total values being more than twice as high as the ambient air VOC concentrations and varying greatly among vehicles. Air samples collected from one of the vehicles showed TVOC concentrations at 7.5 mg/m^3 on the second day from purchase [Grabbs et al., 2000]. The main VOCs identified in the study by Grabbs were toluene in the range of 2.2 - 12.6%, xylene in the range of 2.4 - 10.7%, ethylbenzene in the range of 0.5 - 2.2% and undecane in the range of 0.8 - 7.5% and they showed the same percentage share in the total volume of VOCs detected in the cars. Grabbs et al. [2000] also reported that TVOCs decreased by more than 90% during a three-week test period.

Studies have shown that while the initial concentrations of VOCs are the result of off-gassing of new materials and decrease with time, they are eventually replaced by other compounds that are the result of chemical interactions between various materials present in the interior as well as self-pollution caused by fuel combustion and diffusion of exhaust gases into the interior [Faber et al., 2013b].

Even though older cars also show noticeable amounts of aromatic VOCs such as toluene, ethylbenzene, xylene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, 1,2,3-trimethylbenzene and styrene, and aliphatic VOCs, such as n-undecane, tetradecane, decane, and others, those levels are generally only third of the concentrations of the same compounds found consistently in new cars [Chien, 2007].

Researchers have repeatedly reported that regardless of their actual value, all the pollutants present in new cars record higher concentrations than in older ones [Zhang et al., 2008].

4.3.2 Effect of driving and ventilation mode

Since it has been established that VOCs and other air pollutants accumulate in the car cabin as the result of off-gassing of materials present in the interior, back diffusion of the car's own exhaust fumes and intake of pollutants from the ambient air, it seems understandable that also the driving mode and car ventilation setting can affect the number and total concentrations of VOCs in the car cabin.

By measuring VOC concentrations at various driving (engine off, idling, driving at operating speed) and ventilation scenarios (A/C off, A/C on with re-circulation, A/C on with fresh air intake, windows opened) and their respective combinations it was determined that driving with A/C on re-circulation mode brings the best results in terms of limiting the intake of outdoor pollutants into the car cabin [Chan and Chung, 2003] it may, however, lead to the increase of pollutants in the cabin such as VOC emissions from off-gassing of interior materials, and CO_2. A stationary vehicle with an idling engine turned out to be the worst possible scenario regardless of ventilation mode selected [Kim et al., 2016, Fedoruk and Kerger, 2003, Barnes et al., 2018].

Idling engine mode has the worst effect on VIAQ as it combines the negative effect of self-pollution with the car's own exhaust fumes leaking into the vehicle through the interior, ambient air infusion which takes place even with A/C off and the low efficiency of the car's air ventilation system when the vehicle is not moving. Tests have shown that TVOC concentrations in an idling car can be as much as five times higher than in a parked vehicle with the engine off [Kim et al., 2016].

Kim et al. [2016] found that the concentrations of 17 volatile organic compounds identified inside test vehicles regardless of their age began rising above the ambient air levels immediately as the engine was turned on. Barnes et al. [2018] found that TVOC concentrations increased for the first ten minutes after the engine was switched on and then levelled off and remained even. The VOC levels increase with the engine on regardless of the air ventilation mode selected - interior air re-circulation or exterior air infusion - thus suggesting strong influence of the running engine on VIAQ [Kim et al., 2016].

There can be, however, lower VOC concentrations in a parked car compared to concentrations in an idling car, yet they are still at least twice as high as in the ambient air [Xu et al., 2016] and they are also higher than in a moving vehicle regardless of mode of air exchange applied. Total VOC concentrations in static vehicles mirror off-gassing of materials in the car interior and show negative correlation to increasing interior temperature level [Kim et al., 2016, Brodzik et al., 2014].

Xu et al. [2016] found that with A/C completely off TVOC levels grow to be almost three times the values of ambient air (0.614 mg/m^3), with A/C on re-circulation mode the TVOCs drop to levels only about twice as high as ambient air and with A/C on fresh air intake they spike up again (0.961 mg/m^3) to levels higher than in the first air ventilation scenario.

When switching from A/C with re-circulation mode to A/C with fresh air intake, the concentrations of benzene grew by 29.47%, toluene by 75.43%, xylene by 49.28%, ethylbenezene by 58.26%, styrene by 51.05%, formaldehyde by 20.53%, acetaldehyde by 21.73% and acetone and acrolein together by 104.96% indicating the influence of outdoor pollution and back diffusion of the car's own fumes [Xu et al., 2016].

Since modifications of the ventilation mode brought only a partial improvement to overall VOC in-vehicle-levels which remained significantly higher than ambient air levels, the study results confirmed that the VOC sources are inside the car and the spikes or drops in TVOC levels based on ventilation mode selected were caused by intake of outdoor air.

Fedoruk and Kerger [2003] observed ten-to-twenty-fold lower TVOC concentrations in new cars under driving conditions than under static conditions and four fold lower concentrations in a used vehicle under the same parameters. In another study, 3/4 of vehicles tested under driving conditions in Hong Kong showed TVOC concentrations as good as local standards applied to the interior environment in office buildings. However under idling conditions it was the reverse and 70% of the tested vehicles exceeded those standards by more than five times [Barnes et al., 2018].

Once the car is moving, the selected mode of air ventilation can alter the total VOCs inside only in a minor way. Studies suggest that a relatively high air exchange takes place between the car interior and the ambient air irrespective of the A/C mode selected which can affect only about 20% of the TVOC concentrations. Thus any variance in the VOC levels is a reflection of changes in outdoor air quality more than anything else [Fedoruk and Kerger, 2003].

Similar trends were also observed in a study that focused on the level of infiltration of ultra fine particles (UFP) and carbon dioxide under passive ventilation compared to other different modes of active ventilation [Lee et al., 2015]. The study concluded that outdoor air mode with the fan off (passive ventilation) allowed the highest infiltration of UFP which increased in correlation with increasing speed. Subsequently medium and high fan settings were effective at preventing UFP infiltration at speeds up to 145 km/h.

Except for re-circulation mode, the TVOC levels were higher in all new vehicles which was attributed to better air tightness and absence of cracks and leaks in the cabins of new vehicles which minimized deliberate air exchange between the cabins and the outdoor environment - either by preventing VOCs present inside due to off-gassing of interior materials from escaping or by preventing outdoor polluted air from entering the vehicle [Abi-Esber and El-Fadel, 2013].

4.3.3 Temperature and humidity

As an indoor environment cars are defined as a special micro environment due to the many features that set them apart from what's commonly classified as indoor space, such as office buildings, residential buildings, retail spaces, production halls etc. Besides being characterized by a very small volume of space filled with a high quantity of different materials, it is also defined by high and frequent fluctuations in indoor temperature and humidity which are not always in sync with outdoor conditions.

When parked in direct sunlight, temperature inside a car can approach inhospitable extremes and similarly humidity can temporarily spike in rainy conditions or in humid climates to more than 80% [Geiss et al., 2009, Yoshida and Matsunaga, 2006]. Since glass makes up a large portion of a car interior, compared to a common building, temperatures in a car cabin can spike up quickly and reach close to 90°C with the dashboard heating up to even 120°C [Duh, 2015].

Even with ambient temperature at 30°C, the car interior quickly heats up to 56 - 67°C and even with coolest ambient temperature in the summer the vehicle interior can easily reach temperatures over 47°C [McLaren et al., 2005].

Under normal ambient environment temperature (23 - 30°C) concentrations of VOCs are stable and are not affected by relative humidity, however any deviations from these conditions affect the speed and quantity of material emissions and the total VOC concentrations in the car interior [Brodzik et al., 2016]. High temperatures above the normal level increase the off-gassing of compounds with low and medium boiling points that are generally used in materials in the car interior [Geiss et al., 2009].

Their concentrations in the car cabin can be significantly higher in the summer than in winter, compared to the outdoor environment [Yoshida and Matsunaga, 2006]. The difference between off gassing in the winter and summer months is on average 36 - 42% [Geiss et al., 2009]. In connection to this aspect, it is not surprising that the presence of a sunroof can increase TVOC concentrations by 1.6 times [Brodzik et al., 2014] and should be avoided.

Warm weather and a static vehicle represent the worst combination of conditions that leads to a rapid increase in VOC emissions inside the vehicle. Fedoruk and Kerger [2003] measured that under any combination of driving and ventilation mode the VOC emissions were 20 to 30 times lower than in a static vehicle in warm weather [Yoshida and Matsunaga, 2006].

According to studies focused specifically on classification of volatile compounds and their total concentrations under changing temperatures in a vehicle interior, the total volatile organic compound concentrations increase with every increment of temperature increase and the growth is exponential. The increase in VOC concentrations is not linear and is greater at higher temperatures [Xiong et al., 2015].

It was measured that a temperature increase of ten degrees from 20°C to 30°C brings a 63.3% growth in TVOC, an increase from 30°C to 40°C causes another 42.7% growth of TVOC concentrations and an increase from 40°C to 50°C translates into a 58.5% spike in TVOC values. Also the higher the temperature, the more different compounds can be observed inside a vehicle [Faber et al., 2012].

Due to their different boiling points, not all VOCs respond to rising temperature the same way. Xiong et al. [2015] noted that a five degree point temperature increase to 29°C leads to a 28.8% growth in benzene concentrations, however a further six degree increase (35°C) resulted in a 102% spike of benzene concentrations.

Similar percentage growth was observed for other target VOCs when for example toluene grew by 19% and 103% between 25 - 29°C and 29 - 35°C respectively and formaldehyde by 4% and 117% for the same temperature increments.

Despite the sizeable differences in their concentrations at different temperatures, all of the target VOCs (toluene, benzene, ethylbenzene, styrene, xylene, acetaldehyde and acraldehyde) stayed under the limits defined by the Chinese standard for VIAQ (Code: GB/T 27630-2011) except for formaldehyde which reached peak concentrations of 172.8 - 251.6 g/m^3 at 35°C thus exceeding the limit of 100 g/m^3 more than two-fold [Xiong et al., 2015].

The influence of temperature growth on the TVOC concentrations released was observed also by Fedoruk and Kerger [2003] who measured a nearly five-to-seven-fold increase of VOC concentrations under ”extreme heat” temperatures of 43 - 62°C compared to ”moderate heat” temperatures of 32 - 42°C. Even though the actual concentrations measured for individual cars differed, the trend observed was the same for all the tested vehicles regardless of their age. Newer vehicles, however, showed higher absolute increase and reached higher values. All the vehicles tested showed dominant concentrations of styrene and phenol which are considered clear signs of off gassing of interior materials.

The temperature affects not only the total concentrations of VOCs but also the volume of individual compounds represented in the TVOC. Geiss et al. [2009] observed seasonal variations between the concentrations of carbonyl compounds (acetaldehyde, formaldehyde, propanal, hexanal, acetone etc.) measured in the summer and in the winter. The carbonyl compounds showed on average 42% lower concentrations in December than in August while all the other VOCs showed 36% lower concentrations for the same measuring periods.

Similarly Yoshida and Matsunaga [2006] noted that concentrations of aliphatic hydrocarbons (alkanes and cycloalkanes) in brand new cars decreased rapidly in the first 180 days after delivery, reached about the same levels as the ambient environment during the winter months however spiked again in the summer and exceeded outdoor levels.

On the other hand the concentrations of aromatic hydrocarbons, another group of VOCs that upon delivery recorded concentrations much higher than in the ambient air, decreased after the first 180 days and in the summer were approximately one-tenth to one-hundredth of the initial concentrations. From the second year on there was very little difference in summer and winter concentrations and the concentrations of most of the compounds (except for styrene) were similar to ambient air levels.

Other compounds such as halocarbons, terpens, esters, carbonyl compounds and phthalates, which accounted for less than 10% of the total organic compounds identified in the car interior, showed very minimal seasonal variations and due to their volume share their overall effect on VIAQ can be considered negligible. More so since most of these groups were recorded at levels similar to the ambient air concentrations [Yoshida and Matsunaga, 2006].

Faber et al. [2012] observed that aliphatic hydrocarbons seem to show the biggest growth with a temperature change out of all the different VOC groups generally detected in a car interior, leaping by 341% (or from 0,833mg/m^3 to 2,841 mg/m^3) between the temperatures of 20°C and 50°C with the biggest increment growth (about 42%) being between 40°C and 50°C. A similar trend was observed by Yang et al. [2020] who tested four individual car components and at temperature of 60°C aliphatics made up the majority of all VOCs detected.

Concentrations of aromatic hydrocarbons such as toluene, ethylbenzene, xylene, styrene and naphthalene grew eleven times between 20°C and 50°C, changing their total concentration from 0,110 mg/m^3 to 1,218 mg/m^3. Alcohols, ketones, glycoles and aldehydes showed and exponential growth as well (10, 2.4, 7 and 55 times respectively), however their total concentrations stayed very low and below any exposure limits [Faber et al., 2012].

The change in the composition of emissions is explained by the continuous decline of residual VOCs affixed in the materials. The increased temperature enhances off-gassing of materials due to elevated molecular desorption and molecular speed which subsequently increases the diffusion coefficient. Higher temperatures also encourage emissions of compounds that are in deeper layers of the materials which are often composites [Yang et al., 2020]. For example, the polyurethane foam which is used as padding in seats and upholstery, or adhesives that hold components together on the inside. The issue of adsorption, VOC emissions by multi-layer materials and interactions among compounds in the same chamber, are further discussed in Chapter 4.3.4.

Another factor contributing to the volatile organic compounds emissions is the fact that most materials commonly used in a car interior have a porous structure with the pores serving as potential adsorption sites. According to the Langmuir adsorption theory this means that the level of humidity can affect the quantity of VOC molecules dispersed in the car cabin [Yang et al., 2017, Liu et al., 2015, Czepirski et al., 2000].

As the number of adsorption sites is naturally limited, VOC molecules have to compete with H2O molecules for adsorption. Therefore when relative humidity increases, more water molecules compete with VOC molecules for the limited adsorption sites, the VOC molecules that were adsorbed loose their adsorption position and are released into the air, i.e. the VOC emission rate increases with increased humidity [Yang et al., 2017].

4.3.4 Interactions of materials and compounds

As it has been already established, emissions of volatile organic compounds in a car interior are predominantly caused by off-gassing of materials that make up the components placed in a car cabin. Total VOC concentrations in the car interior however cannot be derived as a simple sum of each of the material's emissions but are rather the result of complex interactions of all of the materials with each other [Brodzik et al., 2016].

It has been established that the presence of certain types of materials can lower the TVOC concentration that should otherwise be higher. Studies have shown that materials placed in the same chamber act as sorbents for chemical compounds emitted by other materials and that the rate of desorption of certain VOCs is slower with specific materials present in the same space [Brodzik et al., 2016, Huang et al., 2006].

The effect of material adsorption and desorption has been primarily observed and studied with construction materials as indoor air quality is generally better monitored and there are limits set for air pollutant concentrations [World Health Organisation, 2010].

Studies of building materials found that adsorption of VOCs onto the materials affects the mobility of indoor contaminants and therefore building materials can have a long-term effect on interior air quality. At first, adsorption can reduce peak VOC concentrations in the room with indoor materials acting as buffers, however, the adsorbed compounds are later re-emitted into the air which can lead to increased TVOC levels in the room [Huang et al., 2006, Meininghaus et al., 2000].

This observation was for example supported in a study by Haghighat and Huang [2003] who found that multi-layer material has a much longer VOC emission time and slower VOC decay rate than a single-layer material. The study showed that a multi-layer material first emits the type of compounds (and VOCs) that reflect the material in the top layer. At the same time, the top layer material can significantly slow down VOC emissions from materials in the lower layers.

The study also showed a sink effect of other materials in the chamber (mainly porous material) on slowing down the initial onset of VOC concentrations; they however elevate with time [Haghighat and Huang, 2003]. This effect of multilayer materials on VOC emissions has been observed also in vehicles [Yang et al., 2020].

This phenomena can be explained by mass transfer mechanisms that have also been observed during experiments measuring the effect of time on VOC concentrations in a car interior. At the beginning the material emissions are driven by convective mass transfer from the material surface [Yang et al., 2020]. As the concentration of VOCs on the surface decreases, the amount of VOCs in the car interior starts to get affected by the diffusion of volatile compounds that did not leave the cabin via air circulation but remained and were adsorbed by other surfaces in the vehicle.

Even though this adsorption and desorption mechanism of VOCs in a closed environment is known, only a handful of studies focusing on this phenomena in the car interior have been carried out so far. This may be due to the fact that VOC emissions in a car are the result of a large number of different sources which makes identifying and measuring them very difficult and also subsequent calculations of their mutual affects require very complicated mathematical models [Jørgensen et al., 2000, Huang et al., 2006].

Adsorption is generally described as a surface process defined by the separation of a substance from a fluid bulk and the accumulation of the substance onto a solid surface. The adsorption can take place using physical forces or chemical bonds. It is usually reversible and the reverse process is called desorption [Artioli, 2008]. The equilibrium of the process is described by an equation of the amount of substance attached to the surface given the concentration in the fluid at a constant temperature and is called adsorption isotherm. There are many definitions describing these equations, the most commonly used are the Langmuir and the Freundlich equations [Huang et al., 2006].

Furthermore adsorption isotherms can be divided into five types that are described based on the type of material they are characteristic of. Based on this classification, type I and type II are typical for non-porous materials characterized by mono-layer adsorption and multi-layer adsorption respectively. Type III is a transitional isotherm where initially there is a little adsorption however once some of the compounds adsorb, more adsorption quickly follows because of strong adsorption relations. Type IV and V are typical for porous materials when the adsorbed layer fills the pores in a rapid and abundant fashion [Masel, 1996, Huang et al., 2006].

As components in the car cabin are mostly made out of materials with porous structure [Yang et al., 2017], it is clear that they must engage in adsorption processes described by the type IV and V adsorption isotherms. Polyvinyl chloride (PVC), nylon, textiles and textile compound materials have all been proven as having a very strong sorption capacity which allows them to act as storage media and a reversible interim store for chemical substances [Jørgensen et al., 2000, Huang et al., 2006, Brodzik et al., 2016] [Bauhof and Wensing, 2008] ex. Ehrler et al. (1994).

The fact that materials in a car interior interact with each other and that their emissions are influenced by the surrounding materials which may in some cases act as sorbents has been proven by experiments studying this phenomena. For instance Brodzik et al. [2016] tested three typical car components - hand break lever cover, sun visor and headlining - to see if the concentrations and the count of the VOCs emitted by these components changed when they were placed next to each other as opposed to when they were tested separately.

The experiments showed that the count of organic substances emitted into the environment was the highest when each part was tested separately and gradually decreased with each part added to the group. Brodzik detected a total of 377 volatile compounds when testing each part separately and then all three of them combined. There were 142 compounds detected in single part testing and 69 organic compounds were present in air samples for all the tests. The tests clearly indicated that the types and the count of the organic compounds changed with each part present or missing in the environment.

The same profound influence of one part on another was established when measuring the concentrations of individual VOCs which confirmed that the level of VOC emissions changed depending on what compounds were in proximity of one another. For example while the hand break lever cover (made out of polyvinyl chloride - PVC) emitted more than 2.5 mg/m^3 of alcohols on its own, when grouped together with the sun visor (made out of hard plastic covered with PVC) and the headlining (made out of polyurethane foam sandwiched in between two layers of synthetic fibers (SF/PUR/SF)) the total alcohol emissions dropped to 1.5 mg/m^3.

Similarly the sun visor emitted extremely high concentrations of aromatic hydrocarbons (2.5 mg/m^3), ethers (3.2 mg/m^3) and hydrocarbons (nearly 3 mg/m^3) compared to the other two parts when tested on its own, however the total concentrations of these organic compounds dropped significantly when also the hand break lever cover and the headlining were present showing about 0.5 mg/m^3 in the case of aromatic hydrocarbons, 0.75 mg/m^3 in the case of ethers and 2.2 mg/m^3 for hydrocarbons.

When comparing total VOC concentrations for each part separately with their total VOC concentration when placed together, it was clear that the materials interact together and influence each other as the total VOC was not a simple sum of the TVOC of the individual parts tested separately. While the total VOC concentration for the hand break cover was 4.75 mg/m^3, 4.72 mg/m^3 for the headlining and over 13.2 mg/m^3 for the sun visor, when placed together their total VOCs reached only 8.2 mg/m^3.

Brodzik et al. [2016] concluded that it was the different materials the components are made out of and the difference in their porosity that influenced the final outcome, as well as how they emit and adsorb volatile organic compounds when in proximity of each other. Materials, such as the headlining, that can be defined by significant porosity, act like sorbents and can bind certain compounds or group of compounds and that way decrease their concentration in the environment.

The adsorption of organic compounds by certain materials can on one hand lead to temporarily decreased VOC concentrations, on the other hand it can prolong the exposure to these substances as based on the definition of adsorption it can be expected that the compounds will be desorbed in the future [Huang et al., 2006, Brodzik et al., 2016].

Even though it focused on materials commonly used in the automobile industry, Brodzik's findings supported similar conclusions established by studies that focused on testing materials used in the construction industry. The phenomena of adsorption of VOCs by different materials used in building construction has been studied extensively as the concentrations of air pollutants in building interiors are closely monitored and regulated unlike in the vehicle interior.

Adsorption and desorption of VOCs on material surfaces has been used to explain the difference between VOC concentrations of single material testing compared to the same material being placed in a room with other materials. Studies indicate that the resulting differences in concentrations are caused by interactions between VOC sources, sinks (materials that adsorb), individual activity patterns of the compounds and ventilation strategies in real rooms [Jørgensen, 2007]. However, similarly to the car environment, studies estimating the sorption effect of material surfaces during real conditions in buildings do not exist.

4.4 Analytical methods to measure VIAQ

The main methods currently used to measure volatile organic compounds in a vehicle interior air are the Gas Chromatography (GC), Gas Chromatography - Mass Spectrometry (GC-MS), High Performance Liquid Chromatography (HPLC) and Selected Ion Flow Tube Mass Spectrometry (SIFT-MS). The GC, GC-MS and HPLC are the industry standards as they have been around for decades and are used for most of the VIAQ testing whether by researchers or by the automobile industry. The SIFT-MS method, which was developed fairly recently however, can offer certain advantages over the other methods; primarily the option of online and real time testing [Smith et al., 2002].

Gas chromatography is a separation technique first described by A.T. James and A.J.P. Martin in 1952 and then quickly adopted by the industry as a reliable, inexpensive and versatile method to analyse volatile materials [Stauffer et al., 2008, Bartle and Myers, 2002].

GC allows for a fast and simple separation of highly complex mixtures based on their different boiling points, vapour pressure and polarity [Stauffer et al., 2008, McNair et al., 2019]. It is applied to organic and inorganic compounds in the form of gases, liquids and solids while the latter usually need to be first dissolved in a volatile solvent [McNair et al., 2019]. GC is valued for separating and analysing multi component mixtures and identifying isomeric forms of compounds mainly in aromatic and aliphatic hydrocarbons [Smith et al., 2002].

While the core analytical method remains the same, there are different techniques to prepare samples for testing or to improve the outcome for certain types of compounds. For example thermal desorption (TD) is a method used to prepare samples prior to inserting them in the gas chromatograph.

During the thermal desorption process the material is heated to better release adsorbed compounds and it is used as a pre-concentration technique mainly for low-concentration analytes such as volatile and semi-volatile compounds that would otherwise be difficult or impossible to detect [MARKES Int., 2017].This technique is used in almost all cases of VOC testing in vehicles and remains the leading technique given the nature of the organic compounds present in the interior air [Widdowson, 2019].

Another commonly used technique is the flame ionisation detection (FID) method which is used together with TD-GC and the mass spectrometry. The TD-GC/FID-MS uses flame ionisation detection (FID) to detect carbon in samples. During the process samples are burned in a hydrogen-air flame to produce carbon ions. Even though the overall process is not very efficient, the total amount of ions is directly proportional to the amount of carbon in the sample [JoVE, 2020].

Mass spectrometry, which can be used to complement gas chromatography, is an analytical method that measures the mass to ratio of ions generated from inorganic or organic compounds and separated by their mass to charge ratio [Gross, 2006]. A mass spectrometer consists of an ion source which is necessary to ionise the analyte (thermally, by electric fields or by impacting charged particles with each other), a mass analyser and a detector, all operated under high vacuum conditions [Gross, 2006].

Gas chromatography used in combination with mass spectrometry is a selective analytical technique that uses chromatography and mass resolution for the analysis of organic compounds [Gröger et al., 2019]. In studies researched for this work TD-GC/FID-MS was the method most commonly used by researchers to analyse VIAQ of tested vehicles or car components.

Another method commonly used to analyse compounds is the high performance liquid chromatography (HPLC) which is a form of liquid chromatography that uses stationary phases consisting of small particles resulting in more efficient separation than the standard liquid chromatography [Moreno-Arribas, 2003]. Since its introduction in the 1960's, HPLC has become an established technique with a great number of stationary phases available [Smith, 2000] and is widely used in medicine, biology and other fields that require accurate and sensitive testing of organic compounds [Itoh and Yamada, 2000, Varma-Basil and Bose, 2019].

The SIFT-MS method is a newer method that, although also using mass spectrometry, can provide real-time analysis of volatile organic compounds [Španel and Smith, 2013, Smith and Španěl, 2005]. The SIFT-MS is a fast analytical technique that works by controlling the chemical ionisation of target gases [Spanel and Smith, 1997] using chemical ionisation of trace gas molecules in air samples by means of helium carrier gas with H_3O^+, NO^+ and $O_2{}^+$ precursor ions to enable analyte specificity [Smith and Španěl, 2005, Kharbach et al., 2018]. Reactions between the precursor ions and trace gas molecules proceed for an accurately defined time. The precursor and product ions are detected and counted by a downstream mass spectrometer which allows for effective quantification.

The SIFT-MS method allows to analyse trace gases without the need to collect them and provides real-time analysis down to parts-per-billion by volume [Španěl and Smith, 2013]. Since SIFT-MS uses soft chemical ionisation that does not require chromatography, it is being marketed by companies that specialise in automotive quality related testing as an easy-to-use instrument for a quick and real-time analysis that can be set up and operated even by staff without technical background [SYFT, 2020]. Since each technique has different fundamental strengths for analysing VIAQ, the two techniques complement rather than replace each other [QUANTUM ANALYTICS, 2020].

4.5 Legislation in regards to VIAQ

4.5.1 Introduction

Legislation and legislative norms are a big part of the VIAQ issue as cars, just like any other product intended for use by the general public, need to comply with a vast number of legislative regulations and requirements. However, while many aspects of the car production and operation are regulated - from car exhaust fumes emissions [Greening, 2001, Rexeis and Hausberger, 2009, Christensen and Gulbrandsen, 2007] to car passengers safety [Moravčík and Jaśkiewicz, 2018, O'Neill, 2009] - the quality of the vehicle interior air goes so far unregulated in most countries.

As was mentioned earlier in this paper, countries in South-East Asia have taken the lead in implementing VIAQ standards for passenger vehicles and therefore have effectively pushed international auto makers to start paying serious attention to the issue.

Car manufacturers have been aware of the issue of excess VOC emissions since the 1970's [Shea, 1971] when concerns regarding VOC emissions were raised with indoor air in buildings and the term "Sick Building Syndrome" was coined [Redlich et al., 1997]. As VOC exposure symptoms are similar whether they are experienced in an office building [Wargocki et al., 1999] or in a car [Faber and Brodzik, 2017, Brodzik et al., 2016], the same concerns that plagued new buildings were mirrored by the auto industry.

The issue of VOC emissions is defined by two areas of concern - one area is represented by the potential threats posed by most VOCs to human health and the other is the olfactory experience of organic compounds' emissions in the car interior. Asian customers are quite sensitive to the "new car smell" and it is among their top complaints with new cars [MARKES Int., 2019]. It is also the reason why Asian countries were among the first to start exploring the issue of VOCs in a car interior.

While health concerns are the primary driver behind the push for industry regulations on the United Nations Economic Commission for Europe (UNECE) level, international as well as Asian domestic car producers are working hard to please the sensitive noses of Asian customers. What further complicates the issue is the fact that lowering VOC emissions to levels where they might not cause adverse health effects does not mean the elimination of unpleasant odors and vice versa [Widdowson, 2019]. The issue of odors is so important for car makers wanting to sell on Asian markets that they have dedicated olfactory teams that study facial expressions to determine the level of pleasantness or unpleasantness of odors [Widdowson, 2017].

According to an automotive quality study by J.D.POWER [2020] comparing the new-vehicle quality gap between Chinese domestic brands and international brands, "unpleasant interior smell or odor" was the most frequent complaint by Chinese consumers and was reported by every 16.4 customers per 100 vehicles. Unpleasant odor ranked in the top 20 quality problems together with eight other sensory issues which concerned mainly excessive and annoying noises such as road noise, wind and break noise, seat squeaks or rattles and others.

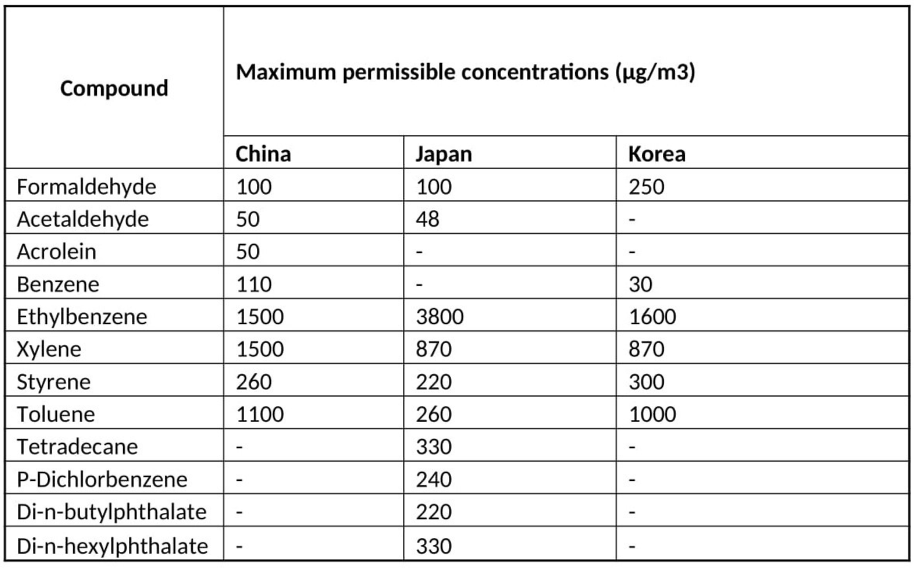
4.5.2 Legislative overview and current situation

There are only a handful of countries in the world that have so far implemented legislation aimed at regulating the levels of VOC concentrations in new cars: China, South Korea, Japan and Russia. In the rest of the world the VOC pollution in car interiors is completely unregulated and limiting harmful emissions from interior components and trim has been left up to the car manufacturers and their own voluntary standards [MARKES Int., 2020].

In 2004 Russia became the first of the four countries to implement test methods and regulations concerning VOCs in vehicle cabins even though the legislation was aimed mainly at curbing pollution from exhaust fumes that can enter the car while in motion. The next was Japan where the Japan Automobile Manufacturers Association (JAMA) in 2005 published a voluntary set of "Guidelines for Reducing Vehicle Cabin VOC Concentration Levels" setting concentration limits for 13 VOCs [UL, 2020].

In 2007 Korea became the first country to implement whole vehicle VIAQ requirements setting emission limits for seven VOCS: benzene, toluene, formaldehyde, ethyl benzene, styrene, xylene and acrolein. The "Newly manufactured vehicle indoor air quality management standard," notification No. 2007-539, issued by Korea's Ministry of Land, Infrastructure and Transportation, also prescribed specific testing methods including vehicle preparation and sampling duration [UL, 2020].

China released its first set of guidelines for VOC emissions permissible in car cabins in 2011 in the national standard GB/T 27630-2011. The "Guidelines for air quality assessment of passenger vehicles" issued by China's Ministry of Environmental Protection and State Administration of Quality Supervision, Inspection and Quarantine came in effect in March 2012 and prescribed concentration limits for the same eight VOCs that were already defined by Korea. Comparison of selected VOC limits for China, Japan and Korea are listed in Table no.2.



Compound	**Maximum permissible concentrations (µg/m3)**		
	China	**Japan**	**Korea**
Formaldehyde	100	100	250
Acetaldehyde	50	48	-
Acrolein	50	-	-
Benzene	110	-	30
Ethylbenzene	1500	3800	1600
Xylene	1500	870	870
Styrene	260	220	300
Toluene	1100	260	1000
Tetradecane	-	330	-
P-Dichlorbenzene	-	240	-
Di-n-butylphthalate	-	220	-
Di-n-hexylphthalate	-	330	-

Table no. 2 – Maximum permissible concentrations of selected VOCs in vehicle interiors by countries

The voluntary guidelines were later revised to become the mandatory national standard and are currently the most stringent rules governing car VOC emissions worldwide [MARKES Int., 2020]. Foreign car manufacturers were given a grace period until the beginning of 2020, however due to the reorganization of Chinese government agencies, automobile makers could expect a further delay past the already postponed deadline of July 2020 [MARKES Int., 2019].

Given the sheer size of the Chinese automobile market (which only in 2019 added 25.8 million new passenger vehicles) [Pham, 2020], it is understandable that all major car manufacturers are closely following the actions of Chinese officials in this matter and try to set their own internal standards in accordance with the Chinese regulations [MARKES Int., 2020].

4.5.3 Norms and methods used for testing VIAQ

Just as important as the emission limits, however, are the methods in place to determine the actual VOC concentrations. Since China is currently the world leader in setting limits for permissible VOC emissions in car cabins, the norms the Chinese regulatory bodies choose to apply will be the ones the automobile manufacturers will largely adopt. And the same goes for testing standards and methods [MARKES Int., 2019].

As car manufacturers have been trying to deal with the issue of VOC emissions on their own, they have each devised their own procedures and methods for testing VOCs and SVOCs in car interiors. This has resulted in hundreds of manufacturer-specific methods producing results that cannot be meaningfully compared with each other [MARKES Int., 2020].

In order to meet the growing demand for industry standards regarding VOC testing of a car interior as a whole, the International Organization for Standardization (ISO) began working on a set of norms that would unite the standards for sampling and analysis of VOCs and SVOCs from vehicle cabins and materials used in their components [MARKES Int., 2020].

The first set of the methods were released in 2012 as ISO 12219, at the same time as the Chinese released their GB/T 27630 regulation demanding their own testing standard method - HJ/T 400. Even though ISO 12219 does not prescribe concentration limits for individual VOCs, it was expected that it would help minimize differences between individual national standards and aide car manufacturers in finding common ground [UL, 2020].

Simultaneously with these efforts, Korea in 2013 submitted a proposal with the United Nations Economic Commission for Europe (UNECE) to examine the matter of VOC emissions in car cabins as it had recorded an increased number of reports by car passengers complaining of headaches, dizziness and other symptoms, all thought to be linked to emissions from materials used in car interiors [GlobalAutoRegs, 2020a]. In response to Korea's proposal, the UNECE established an informal working group to review the issue of VIAQ in November 2014.

The World Forum for Harmonization of Vehicle Regulations (also known as the Working Party 29 or WP.29) oversees three international agreements through which regulators agree on requirements for motor-vehicle safety, environmental impact, fuel and energy consumption, and vehicle-related theft prevention: the

1958 Agreement, the 1998 Global Agreement and the 1997 Agreement Periodic Technical Inspections [GlobalAutoRegs, 2020b].

The Forum has six permanent working parties and one of them is the Working Party on Pollution and Energy (GRPE) which has been tasked with collecting information and reviewing existing standards regarding VIAQ in order to make recommendations that would globally harmonize regulations on vehicles.

The conclusions of the WP.29 findings and recommendations have been compiled in a document called *The Mutual Resolution No. 3 (M.R.3) of the 1958 and the 1998 Agreements concerning Vehicle Interior Air Quality (VIAQ)* which continues to be revised and updated prior to its scheduled adoption in November 2020 [VIAQ IWQ, 2020].

The M.R.3 sums up problems with VOC emissions in car interiors, related health hazards, different emission and testing standards applied around the world, and the compliance issues that car manufacturers face as a result. The M.R.3 suggests harmonised test procedures for the measurement of interior air emissions, taking into account existing standards, however, it does not set any limits concerning VOC emission concentrations, citing regional differences in regulatory stringency of legislation [VIAQ IWQ, 2020].

As a result, the proposed testing method is a compromise between the Chinese HJ/T 400 and the ISO 12219 norms. It suggests testing for eight VOCs (formaldehyde, acetaldehyde, benzene, toluene,xylene, ethylbenzene, styrene and acrolein) using the standardised GC–MS or HPLC methods according to the ISO 16000 norm. The testing should measure emissions from interior materials as well as exhaust emissions entering the vehicle cabin.

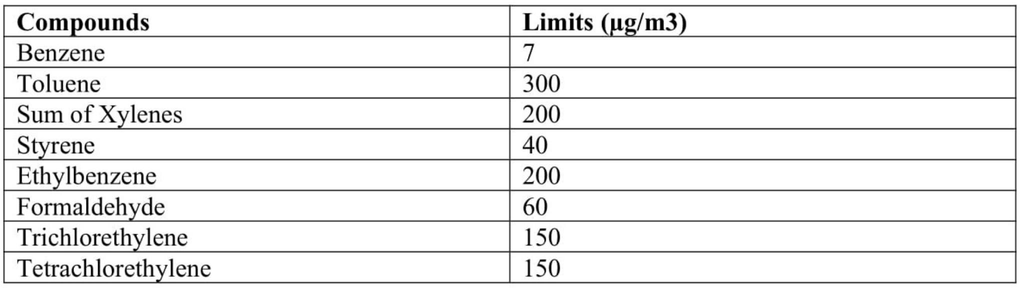
Despite the size of their respective automobile markets, there are no government-led guidelines regarding VIAQ in Europe and the US and all the regulation has so far rested with the auto makers themselves. Some of their industry associations have taken the initiative and made recommendations regarding VIAQ and VOC emissions from materials used in the car interior, for example the SAE International (Society of Automotive Engineers) and the United States Council for Automotive Research (USCAR) in the USA, the Verband der Automobilindustrie (VDA) in Germany or the European Automobile Manufacturers' Association (ACEA) in Europe [MARKES Int., 2017].

4.5.4 Legislative norms regarding interior air in the Czech Republic

There is currently no legislation regulating VOC emissions in a car interior or VIAQ in place in the Czech Republic. The quality of air in indoor environments is defined in Notice no.6/2003 Coll. *Which sets hygiene limits of chemical, physical and biological indicators for the interior environment of specific rooms within certain buildings* described in § 13 of Act No. 258/2000 Coll.*About the protection of public health.*

The specific indoor environments named by the Act are for example school buildings, hospitals, accommodation facilities, retail space and buildings that allow concentrations of large numbers of people, however cars nor any other transportation vehicles are not listed.

The VOCs currently listed in the Notice no.6/2003 Coll. include benzene, toluene, styrene, ethylbenezen, formaldehyde, xylenes, trichlorethylene and tetrachlorethylene. Overview of the regulated compounds and their limits are listed in Table no.3.



Compounds	Limits (µg/m3)
Benzene	7
Toluene	300
Sum of Xylenes	200
Styrene	40
Ethylbenzene	200
Formaldehyde	60
Trichlorethylene	150
Tetrachlorethylene	150

Table no. 3 – Selected VOCs regulated by Notice No. 6/2003 Coll. and their limits for specific interior environments such as residential and office buildings, schools, hotels etc.

4.6 Approaches to improve VIAQ

Taking into consideration all the contributing factors and elements that are responsible for the VIAQ, and mainly the presence of the volatile organic compounds, it is clear that their complete elimination is impossible Johansson [1999]. With regards to vehicle safety requirements and passenger comfort demands it will be impossible to completely abandon the use of materials that emit VOCs in the foreseeable future even though emphasis on their air-polluting aspect could lead to the production of lower VOC-emitting materials by making changes to their chemical composition.

In the mean time research has focused on finding ways to improve air quality in the car cabin by using other methods and means besides the obvious improvement of the material composition of car components.

One way to improve air quality inside a vehicle is to use cabin air filters that are capable of filtering not only the air coming from the outside but also the air re-circulating inside the cabin. Currently, there are four types of car cabin filters used by car manufacturers [MZW, 2020]:

a) **Particulate filter** - it is made out of a porous fibrous material and can trap particles 0.3 microns and larger, such as dust, pollen and other debris.

b) **Charcoal filter** - utilises a normal porous material as the main medium and has a layer of charcoal added to absorb microscopic contaminants. It catches large particles as well as gaseous pollutants such as smoke, exhaust fumes and some VOCs.

c) **Activated carbon filter** - it is made out of a conventional filtration medium and a layer of activated carbon or charcoal. Activated carbon is a type of charcoal produced by heat-treating charcoal and impregnating it with certain chemicals that make it more efficient at eliminating gaseous pollutants such as carbon monoxide, VOCs, bacteria and bio aerosols.

d) **Electrostatic filter** - this type of filter uses a static charge to trap pollutants and works by using layers of electrostatically charged fibers to attract pollutants. These filters are effective at removing particulate matter, dust, pollen, smoke, fumes, spores as well as odors and bacteria.

The most common cabin filters are generally made out of pleated fiber or activated carbon and help reduce the passengers exposure to common air pollutants including bio aerosols such as pollen [Hurley et al., 2019].

Researchers have been looking at ways to increase the efficiency of car cabin air filters in trapping volatile organic compounds by improving materials they are made out of [Moon et al., 2014] as well as experimenting with other techniques such as photo-catalytic oxidation (PCO). The research concerning PCO has focused on titanium dioxide TiO_2 in particular because of its photocatalytical properties that show remarkable results in eliminating VOCs in a laboratory environment [Jo et al., 2002].

In broader terms, PCO is part of the advanced oxidation processes (AOP) that also include photolysis and ozonation, and is recognized as an efficient method that has demonstrated its ability to remove toxic VOCs from indoor environments.

During photo catalytic oxidation, various chemicals are reduced or oxidized on the surface of a photo-responsive material when exposed to a sufficiently high irradiation energy source [Lim et al., 2009]. It is chemically stable, non-toxic and can be used at room temperature to remove many organic compounds under light irradiation [Huang et al., 2016]. Also compared to other known methods, such as bio-filtration, adsorption and thermal catalysis, it is very cost effective [Lin et al., 2013].

Photo-catalytic reactions can be carried out in the presence of photo-catalysts. Those are solids that encourage reactions in the presence of light but are not themselves consumed in the reaction. There are many known compounds that can serve as photo-catalysts in PCO reactions as they have sufficient band-gap energies necessary to promote photo-catalytic reactions, for example Fe_2O_3, SnO_2, TiO_2, CdS, ZnO and ZnS. Out of these semiconductors, TiO_2 molecules are regarded as the most promising nano-structures to serve as photo-catalysts since they promote oxidation of most VOC pollutants at room temperature and don't require any chemical additives [Lin et al., 2013].

Other technologies used for the removal of VOCs include adsorption, liquid absorption, catalytic combustion and membrane separation. Many of these techniques have been applied by the industrial sector but they still need to be further developed and optimized for smaller scale use.

For example adsorption uses porous materials such as activated carbon, molecular sieve or silica gel to physically and chemically adsorb pollutants. However while adsorption works well with high water solubility VOCs such as formaldehyde, other adsorbed gases could be re-released should conditions such as temperature and relative humidity change, which is of concern [Huang et al., 2016].

The use of TiO_2 as a photo-catalyst to reduce VOCs in air has been successfully tested for VOC concentrations in the low to high parts per million (ppm), which are typical for industrial chemical processes, as well as concentrations in the low parts per billion ($<$ 100ppb), typical for VIAQ [Jo et al., 2002].

Titanium dioxide has been extensively studied due to its superior photo-catalytic oxidation ability, excellent stability, high photo-corrosion resistance, low cost and the fact that it is non-toxic. The only downside of using TiO_2 is that due to its large band gap it can be activated only by ultraviolet light (UV) which makes up only 5% of sun light. However experiments have been carried out using visible light for the removal of toluene, xylene and acetaldehyde as well and showed a 90% success rate.

Generally the best results with TiO_2 as a photo-catalyst are obtained when a UV lamp is used for the process [Huang et al., 2016]. When tested in laboratory conditions with a UV light source the removal of common volatile organic compounds such as benzene, ethyl benzene and xylene compounds was nearly 100% [Jo et al., 2002].

Experiments carried out by the Korean Railroad Research Institute aimed at reducing VOC concentrations and bacterial pollution inside passenger railroad cars demonstrated that materials coated with TiO_2 layer emitted less VOCs even when UV lamps were not applied and showed a decrease with time compared to non-TiO_2 coated materials whose VOC concentrations rose sharply and remained at a steady level [Cho et al., 2015]. Under UV lamp light VOC concentrations shot up immediately for both coated and non-coated materials, however VOC emissions from the coated material decreased at a higher degree.

Other researchers have experimented with modifying TiO_2 by carbon nanotube (CNT) composites to enhance the photo-catalytic activity under visible light [Wongaree et al., 2016]. This method shows promising results as CNT/TiO_2 nanoparticles can decompose harmful organic compounds to carbon dioxide and water. Even better results are obtained when nanoparticles are weaved into nanofibers as they show good inter-connectivity of pores, are easy to process and can be recovered when used in water and air applications. Especially the issue of their recovery presents a technical barrier for using nano-particles in environmental applications [Sun and Wang, 2014].

Researchers have focused on developing dye-sensitised TiO_2 (metal-doped or nonmetal-doped) as a way to extend its optical absorption to the visible light range as well as using carbon nano-fibers to enlarge specific surface area to increase the amount of light absorbed by TiO_2 [Wongaree et al., 2016]. Other methods include experimenting with non-TiO_2 catalysts that have low band gaps or combining PCO with other processes that encourage degradation efficiency [Huang et al., 2016, Khan and Ghoshal, 2000].

The use of electrospun carbon nanotube titanium dioxide (CNT/TiO_2) nanofibers, for example, showed very good results at removing gaseous benzene via photo-catalytic degradation by removing 52% of the gas within 90 minutes [Wongaree et al., 2016]. The use of CNT created a larger specific surface area compared to plain TiO_2 which helped with adsorption of the compound. The use of nanofibers could make this method suitable for use in filters as they are easy to replace and can be used in combination with carbon filters to remove most of the common air pollutants.

As it was mentioned earlier activated carbon filters are capable of adsorbing volatile organic compounds as well as particulate matter, pollen and other pollutants. Commercially available filters are generally made out of non-woven polypropylene (PP) combined with activated carbon, however due to the existence of macro-pore structure their speed of VOC adsorption is very slow and they usually cannot be recycled [Moon et al., 2014].

Researchers have been experimenting with a new type of filter made out of activated carbon fibers (ACF) that, due to an increased specific surface area and low mass transfer resistance, show adsorption speed of VOCs to be a minimum of 100 times higher than with regular activated carbon filters. While comparing the ability of various types of filters to remove the most common VOCs present in car cabins, such as formaldehyde, benzene and toluene, activated carbon fiber filters showed the best results.

In an experiment carried out by Moon et al. [2014], regular non-woven PP filters removed only 20% of formaldehyde and virtually no benzene or toluene while non-woven PP filters with active carbon removed more than 97.5% of benzene, 99.3% of formaldehyde, however left 78.9% of the initial concentration of toluene. The ACF filters removed more than 97.5% of toluene and benzene and more than 99.3% of formaldehyde. Further experiments showed that the best adsorption capacity was reached with an ACF filter with a pore size of 2 - 5nm.

Activated carbon fiber filters show similarly positive results when tested for use in HVAC (heating, ventilation and air conditioning) units in commercial buildings. ACF filters removed between 50 - 80% of most VOCs emitted into the air by materials used in building interiors, with higher efficiency observed for air circulated through ACF heated to 150 °C [Sidheswaran et al., 2012]. ACF's potential is also supported by its long adsorption life time which can be periodically regenerated.

Similarly encouraging experiments have been carried out with ACF filters calcined with copper oxide catalyst where the copper oxide (CuO) serves as a strong oxidizer [Huang et al., 2010]. By combining the oxidizing and absorbing capabilities, the CuO/ACF catalyst filter showed increased VOC removal efficiency.

4.6.1 Potential of VIAQ modeling

As established earlier, material VOC emissions in car cabins change with temperature as well as with humidity, air flow and vehicle age. Taking these factors into consideration researchers have looked at the possibility to predict VOC concentrations by designing specific analytical mathematical models or by using known VOC emissions from certain types of materials and modeling their combined effect on TVOC.

For example Yang et al. [2017] has proposed a method to determine the influence of environmental factors on the concentration of VOCs found in vehicle cabin using the initial emittable VOC concentration (Co) and the diffusion coefficient (Cm) of VOCs emitted by materials in a vehicle cabin as the key parameters for describing the emission behaviors of material VOCs [Yang et al., 2019].

A similar method is already being used to determine VOC emissions in the indoor environment of buildings under closed and ventilated conditions. However, while this method works with the diffusion coefficient (Dm) and material/air partition coefficient (K), Yang proposed to work with the Co instead which he considers to be the most sensitive key parameter affecting emission behaviors. This method also assumes a certain amount of VOC emissions to be already present in the environment, a situation common under real in-vehicle conditions.

In this method he first established a linear correlation between the logarithm of access VOC concentration and emission time which can then provide the necessary Co and Dm for various VOCs under different environmental conditions. Yang tested the method by comparing in-lab measurements of real VOC emissions under different temperatures with results derived by his proposed method and concluded that the data demonstrated the effectiveness of his method.

Other researchers have focused on finding similarities and patterns in VOC concentrations by comparing the same models by one manufacturer or same models by different manufacturers to establish whether total VOC emissions are consistent and whether they could be predicted based on a known combination of components. However while one study found intra-model variability to be only 14% [Brodzik et al., 2014], another study found it to be more than 47 % [Chien, 2007].

A study by Brodzik et al. [2014] that compared VOC emissions in nine newly manufactured cars of the same brand and model with different interior fittings found that there were differences between cars with different interior fittings however the difference between two identical cars was less than 14%. Brodzik also found that the total number of VOCs was in positive correlation with TVOC concentrations and therefore suggested that total VOC concentrations in a vehicle could be predicted based on the number of compounds detected.

Chien [2007] tested 20 cars by four different manufacturers and found that the mean intra-model variability was 47% while the mean intra-brand variability was 95%. The differences between manufacturers were so great that while cars with leather trim showed increased TVOC concentrations for one manufacturer, for another one they were lower than cars with fabric trim. Chien concluded that the big difference in TVOC concentrations for same models by different manufacturers was caused by using different manufacturing processes, solvents and component suppliers.

Even though the tested vehicles as well as the testing methods varied, it is clear that manufacturers cannot for various reasons maintain the same VOC concentration in their cars even if they are identical. This fact could make any predictions almost impossible.

5 Discussion

In modern society people spend on average 87% of their time indoors and while most of this time they are inside buildings (Jenkins et al., 1992 ex. [Hodgson and Levin, 2003]), a sizeable portion of this time is also spent inside vehicles [Kim et al., 2016]. Therefore the car has become an important micro-environment with an effect on human health [Liang et al., 2019].

Much like occupants of new office buildings in the 1970's who began complaining of a range of health problems from headaches and nausea to asthma and allergies [Hodgson et al., 1994], later recognised as the Sick Building Syndrome [Jansz, 2017], with the prevalence of car transportation car passengers have begun reporting the same problems [Brown and Cheng, 2000]. Both the occupants of the buildings and the car passengers were describing symptoms caused by the same problem - excess emissions of volatile organic compounds in the air.

The most common VOCs found in a car interior are benzene, toluene, ehtylbenzene, xylene, styrene, formaldehyde, naphthalene and phthalates and they all can have negative effects on human health depending on the duration and concentration of exposure. Some are proven carcinogenics and endocrine disruptors and for example benzene does not have a safe exposure level [Smith, 2010].

Yet they are all routinely present in the interior of cars in concentrations that are five to ten times higher than in outdoor air and up to three times higher than in other indoor environments [Faber et al., 2013b] ex. [Weisel, 2005], [Jo and Park, 1999, Xu et al., 2016]. The VOCs have also been found to act as precursors to other air pollutants with negative health effects of their own thus increasing overall health risks to car users [Bolden et al., 2015].

Despite these facts, very little attention has been paid to this issue by the general public and there are hardly any restrictions in place regarding VIAQ anywhere in the world. As the research for this paper has shown, elevated VOC concentrations are a problem common to all cars regardless of their producer, model or price range [Janicka et al., 2010, Zhang et al., 2008].

So far South-East Asia has been the only region that has taken steps towards limiting the VOC presence in vehicles, with China establishing the strictest legally binding limits [UL, 2020]. While health concerns are certainly part of the equation, the initiative of Asian countries has been more likely driven by the heightened sensitivity of Asian people to odors than anything else [MARKES Int., 2019]. In fact the "new car smell" so desired by the Western world [Krishna, 2013], is the number one complaint for Asian customers [J.D.POWER, 2020].

Because of this difference in perception, car users in the rest of the world are at the mercy of auto makers and are left to self-imposed limits by auto manufacturers and their industry associations. In my opinion, it is because especially Europeans and Americans value the new car smell as a proof of their status and achievement and since it doesn't bother them, there is very little awareness among the public about the VOCs in the car cabin and their health effects.

Even though car manufacturers have started paying attention to the issue, largely being forced by China's requirements, there is no sign of wider international legislative changes. The UNECE has moved in 2013 to review international regulations and requirements regarding VIAQ but this was only so it could "harmonise" international standards in regards to testing methods. The process is still under way and when the new resolution is passed (scheduled for sometime in 2020) it will not set any VOC concentrations limits [VIAQ IWQ, 2020]. In the Czech Republic a car interior is not recognised by the legislation as an indoor environment at all which means that air quality in a car is not regulated.

The issue of VIAQ and especially the presence of volatile organic compounds should not be taken lightly because as the research into VIAQ has shown, the number of volatile organic compounds in a car can be over 200 [Yoshida and Matsunaga, 2006] with concentrations exceeding indoor air limits many times over [Grabbs et al., 2000]. Given the inadequate legal protection of car users and the non-existent VIAQ standards in most parts of the world, car users can best protect their own health when aware of factors and situations that affect the increase or decrease of VOC concentrations in the car interior.

For example, one of the strongest factors is vehicle age with the highest number of compounds and their highest concentrations found in new cars just days or weeks after delivery. These figures decrease with time and cars between three to five years of age showed the lowest VOC concentrations [You et al., 2007].

Also temperature and humidity affect VOC emissions that grow with increasing temperature [Geiss et al., 2009, Yoshida and Matsunaga, 2006] and humidity and decrease under good ventilation and during driving [Chien, 2007, Chan and Chung, 2003]. To reach the best VIAQ, a car should be parked in a covered garage (never in direct sunlight) at a temperature between 20 °C to 25 °C [Brodzik et al., 2016] with windows down to ensure air circulation. It should be driven with windows closed and A/C on re-circulation [Xu et al., 2016]. If the traffic is low and the ambient environment is clean, it is possible to drive with windows down or the A/C on fresh air intake.

Aware of these trends, researchers have focused on identifying the main sources of VOCs in a car cabin and established that it is the off-gassing from materials used in the car interior that is responsible for majority of VOC emissions [Faber et al., 2014]. Even though other factors such as self-pollution with the car's own exhaust [Rahman and Kim, 2012, Abi-Esber and El-Fadel, 2013], the intake of polluted ambient air or the use of air fresheners can also affect TVOCs, material emissions represent the most significant and permanent source of VOCs.

As each car component is made of different materials emitting different VOCs, every change to the interior fittings naturally effects overall VOC emissions. For example choosing fabric trim seats over leather can make a big difference as car leather emits more toluene, formaldehyde and acetaldehyde than fabric-covered seats [Xu et al., 2016, Bakhtiari et al., 2018].

Even the choice of a color makes a difference as each change of the interior color scheme is mirrored in changes of VOC types, their count and concentrations [Faber et al., 2014, 2013a]. Testing has shown that cars with lighter colors show lower TVOC concentrations and for example cars with predominantly black upholstery have up to 30% higher TVOC concentrations than cars with at least some white or red trim.

Considering these findings and given the fact that the black color also encourages heat absorption (and higher temperatures increase VOC emissions), it would be wise for car producers to refrain from using black interior fittings, currently the default color scheme in most cars. The same recommendation goes for sunroofs which can increase TVOC concentrations by 1.6 times and therefore should be avoided [Brodzik et al., 2014].

Some researchers have been asking the obvious question whether it would be possible to estimate or even calculate VOC emissions based on the known material composition of a car interior. While some research suggested that intra-model variability for cars with identical equipment could be only about 14%, glitches in the production process could cause a random difference of up to 200% in TVOC between two identical cars [Brodzik et al., 2014, Chien, 2007]. Such unpredictable fluctuations would most definitely rule out the possibility of predicting VOC emissions or even their specific levels to meet certain predefined criteria.

Other researchers tried designing mathematical models that would allow for a calculation of total VOC concentrations based on known emissions from certain materials and other parameters [Yang et al., 2017]. Even if these calculations may have worked in a lab setting, in my opinion they would be impractical and possibly unreliable in real life. As each car manufacturer uses many different materials for each model, it would be very time consuming and costly to create a reference database for every material used during the production process so that the total VOC concentration could be calculated. And finally, the model would have to be adjusted to include tens of input values.

Another factor that complicates any possible predictions is the fact that materials in a closed environment interact with each other and the presence of one material can influence the emissions of VOCs by another material [Haghighat and Huang, 2003]. A phenomena first studied with building materials was confirmed by experiments carried out with common car components [Brodzik et al., 2016]. It was observed that the presence of a certain (usually porous) material can significantly decrease TVOC levels. However such an adsorption is not permanent and experiments have shown that under changed conditions all of these VOCs are later released and may in fact increase TVOC concentrations [Huang et al., 2016].

As one can see even when certain trends and patterns of behavior regarding VOCs can be observed, there are still too many known and unknown factors that influence TVOC concentrations in a particular car. Certainly more research is necessary to determine what materials can be or should be placed together to achieve lower TVOC concentrations and what material or color combinations should be avoided.

In my opinion it is this near impossibility to predict VOC emissions and the fact that so many different aspects can influence the outcome that the international legislative efforts have been so guarded and basically inadequate given the health consequences. Even so, car manufacturers are still responsible for VOC emissions from materials they place in the cabin and besides the obvious improvement of material composition, there are other ways they can take to control VIAQ.

The easiest one would be an installation of improved interior air filters that could actively lower VOCs, such as filters with activated carbon fibers or filters coated with Titanium Dioxide (TiO_2). While TiO_2 coated filters show a near complete elimination of VOCs [Jo et al., 2002], their wider application is limited as they require UV light for best results. However activated carbon fiber filters can be implemented immediately and their successful removal rate is between 50-80% [Sidheswaran et al., 2012].

Another step would be to control VOC emissions at the place of their origin, i.e. in the car factory. It is important that car producers are aware of TVOC concentrations in their cars even before they are completed so that they can replace higher emitting materials or adjust the production process to lower the amount of post-production VOCs.

To do that car producers need to be continuously monitoring VOC content of individual components as well as of whole cabins during the production process. As regular testing requires time consuming and costly lab analysis, car producers could use the SIFT-MS method that seems to be well suited for this purpose. It provides quick, reliable and on-line results and is easy to carry out even by untrained staff [Španel and Smith, 2013]. In my opinion car manufacturers need to be aware of VOC emissions already at the design stage to choose the right kind of materials for the interior.

Another possibility, not explored by this paper, would be to consider replacing some of the man-made materials with natural materials when suitable. For example using sheep wool for seat cushions instead of polyurethane foams. Even though with today's numbers of new cars, car manufacturing can't go back to making cars out of wood and leather, but perhaps there could be other ways to reduce the quantity of artificial materials in the interior.

Based on the close attention car manufacturers are paying to China's regulations and requirements, it can be assumed, that they are already carrying out their own internal VIAQ testing. These results, if made public, would provide customers with valuable information and would give them another tool to help with decision making when choosing a new car.

Once the Chinese legislation regarding VOC limits comes in effect including all cars sold on the Chinese market, all car manufacturers will have to adhere to those standards or loose access to the market which saw nearly 26 million new cars in 2019 [Pham, 2020].

Since car manufacturers will have to monitor and adjust their production process to meet the Chinese standards, it would seem logical to carry those improvements over to the other markets as well and make them part of the car maker's marketing aimed at improving the brand's perception. This on the other hand could turn out to be a double-edged sword for them as it could bring unwanted attention to the in-cabin VOC issue which non-Asian customers are so far blissfully unaware of.

So it will remain to be seen what strategy the car makers will choose. Because just like everything else, better quality usually means a higher price. And with the highly competitive automobile market, the price is almost always everything.

6 Conclusions

The car interior is a very important micro-environment with an effect on human health. It is filled with many man-made materials that all emit volatile organic compounds in a varying degree. As a result the concentration of VOCs in a car interior is several times higher than in outdoor air or other types of indoor environments and is strongly influenced by additional VOC sources such as self-pollution by car's own exhaust fumes, intake of polluted air from the outside, the use of car air fresheners and smoking. All VOCs commonly found in a car interior have negative effects on human health, many of them are proven carcinogenics and endocrine disruptors and combined with other air traffic pollution represent a significant health hazard to all car users.

Despite the contributing influence of exterior factors, materials that make up components in the car interior are by far the biggest source of VOCs in a car cabin. Time since manufacture is the strongest factor, with newest cars showing the highest VOC concentrations as well as the highest number of compounds. VOC concentrations decline with time, with the biggest change observed in the first three years of a vehicle age.

Temperature is the strongest influencer of VOC concentrations regardless of the vehicle age. Total VOC concentrations increase in correlation with temperature and higher temperatures can increase VOC levels many times over even in older vehicles. Factors that can positively influence VOC concentrations are driving and good ventilation, additional factors are type of fuel (effect of back-diffusion) and size of a vehicle (the lower the car the more polluted air it pulls in with its A/C system).

Materials present in the car cabin can influence each other's emissions and total VOC concentrations are affected not just by the material selected but also by its color or the presence of a sun-roof. Higher price of a vehicle does not always guarantee lower VOC emissions even if the materials used are of higher quality, such as leather, since they may be treated with more VOC emitting compounds to ensure better consumer experience.

There is no international standard regarding vehicle interior air quality and only several countries have so far passed legislation setting VOC limits in cars. China has the strictest rules mandating both VOC concentration limits and required testing methods effectively pushing international car makers to pay attention to the issue. The Czech Republic has no restrictions regarding VIAQ.

Since there is a great number of factors that influence VOC emissions in a car, TVOC concentrations can vary even between identical models by the same manufacturer. Ways to improve the VIAQ include improved material composition, car air filters that can actively eliminate VOCs and better consumer awareness about the issue. Because right now the only way car users can protect themselves from high-risk exposure to elevated VOC concentrations is by making educated choices regarding the vehicles they use and the way they use them.

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