

Volatile methacrylates in dental practices

Wolfgang Marquardt^{a*} / Mario Seiss^b / Reinhard Hickel^c / Franz X. Reichl^d

^a Doctoral Student, Department of Operative Dentistry and Periodontology, Ludwig-Maximilians-University, Munich, Germany

^b Research Assistant, Walther-Straub-Institute of Pharmacology and Toxicology, Ludwig-Maximilians-University, Munich, Germany

^c Director, Department of Operative Dentistry and Periodontology, Ludwig-Maximilians-University, Munich, Germany

^d Professor, Walther-Straub-Institute of Pharmacology and Toxicology, Ludwig-Maximilians-University, Munich, Germany

* Corresponding author:

Ludwig-Maximilians-University

Walther-Straub-Institute of Pharmacology and Toxicology

Goethestr. 31

D-80336 Munich

Tel.: +49-(0)89-2180-75753, Fax: +49-(0)89-2180-75701

E-mail address: Wolfgang.Marquardt@lrz.uni-muenchen.de

Abstract

Purpose. In the recent years an increase of occupational respiratory diseases such as asthma caused by methacrylates was observed in dental personnel. In this study the exposure of dental personnel to various volatile methacrylates was investigated.

Materials and Methods. The air levels of methacrylates were measured during filling therapies while bonding agents were used in four dental practices in Munich, Germany. Short-term air sampling (15 min) was performed using solid phase microextraction (SPME). The SPME fibers were coated with carbowax/divinylbenzene to enrich the analytes. For analysis the analytes were thermal desorbed from the fiber and subsequently analyzed directly by gas chromatography-mass spectrometry.

Results. The methacrylates methyl methacrylate (MMA), 2-hydroxyethyl methacrylate (HEMA), ethyleneglycol dimethacrylate (EGDMA), and triethyleneglycol dimethacrylate (TEGDMA) were identified in the air of dental practices. The exposure levels of the four methacrylates varied during the filling therapies. The maximum concentrations found were 0.4 mg/m³ for MMA, 45 µg/m³ for HEMA, 13 µg/m³ for EGDMA, and 45 µg/m³ for TEGDMA. The detection of TEGDMA correlated with the application of bonding agents during dental fillings.

Conclusion. Exposure levels of different methacrylates were observed at all investigated dental practices. The maximum levels of MMA measured in this study were at least 200 times lower than the toxicologically relevant maximum allowable concentrations defined in various countries. Nevertheless, the exposure levels of methacrylates should be kept as low as possible due to the allergenic potential of some methacrylates.

Keywords: Methacrylates; Bonding agents; Air monitoring; Dental practices;
Respiratory diseases

Introduction

Dental personnel are exposed daily to a diverse amount of different chemicals such as methacrylates. Methyl methacrylate (MMA) is a methacrylate which has been used since several decades in dentistry. The monomer MMA is a volatile, flammable, and colourless liquid. It is mixed with pre-polymerized MMA powder (PMMA) to make various dental products.¹⁰ Bottles of liquid monomer containing more than 90 % MMA are used directly in dental practices. Because of its volatility, MMA is released directly into the air.¹⁹

Acrylic-based polymers have been used in restorative dentistry since the early fifties of the last century, though its use increased in the 1990s to replace metallic restorations using materials such as amalgam.^{5,31} Moreover, acrylic-based polymer materials are used in bonding agents and dentures. Bonding agents usually contain 2-hydroxyethyl methacrylate (HEMA) and triethyleneglycol dimethacrylate (TEGDMA).⁵ Other methacrylates used in dental materials, e.g. ethyleneglycol dimethacrylate (EGDMA) as well as HEMA and TEGDMA are less volatile than MMA due to their high boiling point but might be released as well into the air.

Several previous studies investigated the cytotoxicity of methacrylates on different cell culture systems.^{17,24-26} Due to their toxicological relevance a maximum allowable concentration for the volatile MMA in the air at workplaces is set to 210 mg/m³ in Germany and varies between 100 and 400 mg/m³ in other countries.^{3,30}

Further studies revealed allergic risks of some methacrylates. Methacrylates such as MMA and EGDMA are known as strong sensitizers.^{16,21,29} A well-known complaint among dental personnel is contact dermatitis caused by these methacrylates.^{8,12,20} Contact dermatitis is initialized generally by direct contact with a sensitizing methacrylate, but Kanerva *et al.* described as well contact dermatitis initialized by the exposure of skin with air containing a sensitizing methacrylate.¹⁵ Geukens and Goossens identified HEMA, EGDMA, and TEGDMA as responsible for most of occupational contact allergies in dentistry.⁴

Except for contact allergenic reactions of methacrylates occupational respiratory diseases are known among dental personnel. Case reports have been published on occupational respiratory hypersensitivity such as allergic alveolitis, rhinitis, and asthma.^{9,22,23,27} Most of the cases of occupational asthma were caused by inhaled methacrylates,^{22,23} e.g. a severe asthmatic reaction caused by an allergy on MMA lasted for 13 years.¹

The knowledge about the volatile methacrylates released into the air at dentists' workplaces is scarce.⁶ A recent study investigated the exposure levels of MMA and HEMA in dental clinics.⁵

The aim of this study is to investigate the exposure of dental personnel to volatile methacrylates in dental practices during dental filling therapies while bonding agents were used. The solid phase microextraction (SPME) method¹⁸ was used for the sampling of volatile methacrylates in the air and the gas chromatography-mass spectrometry (GC-MS) was used to analyze the samples.

Materials and Methods

Chemicals

Methyl methacrylate (MMA), 2-hydroxyethyl methacrylate (HEMA), ethyleneglycol dimethacrylate (EGDMA), and triethyleneglycol dimethacrylate (TEGDMA) were purchased from Fluka (Buchs, Switzerland) with the highest purity available. Stock solutions were prepared by diluting the methacrylates with water (HPLC grade) and methanol (HPLC grade) purchased from Fluka (Buchs, Switzerland).

Air Sampling

Air sampling was performed during the treatment of patients at four different dental practices in Munich, Germany. The rooms for treatment varied in size between 35 m³ and 50 m³. In general, one dentist, one dental assistant, and one patient were present during the air sampling. Samples were taken directly in the work range of the dentist at five different treatments in each dental practice.

Four individual sampling fibers were used during each treatment one after another to investigate temporary changes of concentrations of the methacrylates during treatment. The sampling time of each fiber was 15 min, therefore the total sampling time was one hour. The first sampling fiber was used directly before the filling therapy on the patient started to investigate concentrations of methacrylates in the air caused by previous therapies and to identify any possible accumulation during measurement. Therefore, every filling therapy started at a sampling time of 15 min and ended during the sampling

time, influenced by e.g. the extension of the treatment or the working method of the dentist. The time frame of 15 min for air sampling stays in line with the short term exposure limit used to identify exposure levels during specific work tasks and allows direct control of the results (e.g. 15 min short term exposure limit for MMA in Germany: 420 mg/m³). All therapies were filling therapies and four practices were chosen randomly to ensure results as ranges of methacrylates concentrations independent of the used materials, the work technique of the dentist, ventilation or other factors.

The sampling was performed with solid phase microextraction (SPME, Supelco, Bellefonte, USA). An optimum enrichment of methacrylates combined with a suitable matrix separation was achieved by choosing carbowax/divinylbenzene fibers (stable flex; film thickness, 70 µm). The sampling time with each fiber was always 15 min. This time was proved to be sufficient to enrich the analytes on the SPME fiber surface under equilibrium conditions. The temperature during the indoor sampling was 22 °C ± 2 °C in all dental practices. The SPME samples were analyzed directly after the sampling.

GC-MS Analysis

After the enrichment of the analytes on the SPME fiber they were thermal desorbed from the fiber surface for 5 min in the injector of the gas chromatograph. The samples were analyzed by gas chromatography-mass spectrometry (GC-MS) on a Finnigan Trace DSQ mass spectrometer (Thermo, Dreieich, Germany) coupled to a Finnigan Trace GC Ultra gas chromatograph (Thermo, Dreieich, Germany).

A VF-5ms FactorFour (Varian, Darmstadt, Germany) capillary column was used for GC (length, 30 m; inner diameter, 0.25 mm; coating, 0.25 μm). Helium was used as carrier gas (flow rate, 1.0 mL/min). The temperature of the split-splitless injector was 220 $^{\circ}\text{C}$. The GC oven was heated from 50 $^{\circ}\text{C}$ (3 min isotherm) to 250 $^{\circ}\text{C}$ (2 min isotherm) with a rate of 50 $^{\circ}\text{C}/\text{min}$.

The temperature of the direct coupling to the mass spectrometer was 250 $^{\circ}\text{C}$. The temperature of the combined EI/CI source was 200 $^{\circ}\text{C}$; the electron energy was 70 eV.

Mass selective detection was performed with either full scan (40–340 amu) for identification or with selected ion monitoring (SIM) mode for quantitative analysis. The qualifying ions for SIM mode were m/z 69, 87, and 100 for MMA and HEMA, m/z 69, 113, and 170 for EGDMA, and m/z 69, 100, and 113 for TEGDMA. SIM mode was used with a dwell time of 33 ms.

Calibration

The GC-MS was calibrated to quantify the identified methacrylates MMA, HEMA, EGDMA, and TEGDMA. The calibration of the GC-MS was performed with headspace SPME analysis in 1.11 L glass flasks sealed with a Teflon-coated septum and filled with different methacrylate concentrations. The sampling of the methacrylates for the calibration was performed in the same manner as the sampling in the dental practices (carbowax/divinylbenzene fibers, 15 min, 22 $^{\circ}\text{C}$). The analysis of the calibration samples was performed with the same GC-MS program used for the air samples received from the dental practices. Multipoint regression curves were calculated from these data points. The calibration range for MMA was 0.04 to 54 mg/m^3 with a

quantification limit of 0.05 mg/m³. The calibration ranges were 5 to 54 µg/m³ for HEMA, 5 to 225 µg/m³ for EGDMA and 9 to 45 µg/m³ for TEGDMA, respectively. The limits of quantification were 6 µg/m³ for HEMA and EGDMA, 13 µg/m³ for TEGDMA.

Statistics

All values were expressed in µg methacrylate / m³ air. The results in the figures were represented with 95 % confidence intervals (95 % CI).

Results

Practice 1

The results for the short-term measurements of methacrylates during composite filling therapies in *Practice 1* are presented in Figure 1. The methacrylates MMA, HEMA, and TEGDMA were detected during treatment. The concentrations for MMA varied from 60 to 100 $\mu\text{g}/\text{m}^3$ (Figure 1a). The exposure levels for HEMA were between 7 and 45 $\mu\text{g}/\text{m}^3$ (Figure 1b). The air levels of TEGDMA varied between 19 and 32 $\mu\text{g}/\text{m}^3$ (Figure 1c).

Practice 2

The results for the short-term measurements of methacrylates during composite filling therapies in *Practice 2* are presented in Figure 2. The methacrylates MMA, HEMA, and TEGDMA were detected during treatment. The concentrations for MMA varied from 60 to 180 $\mu\text{g}/\text{m}^3$ (Figure 2a). The exposure levels for HEMA were between 12 and 20 $\mu\text{g}/\text{m}^3$ (Figure 2b). The air levels of TEGDMA varied between 24 and 39 $\mu\text{g}/\text{m}^3$ (Figure 2c).

Practice 3

The results for the short-term measurements of methacrylates during filling therapies in *Practice 3* are presented in Figure 3. The methacrylates HEMA and TEGDMA were detected during treatment. The exposure levels for HEMA were between 9 and 24

$\mu\text{g}/\text{m}^3$ (Figure 3a). The air levels of TEGDMA varied between 29 and 45 $\mu\text{g}/\text{m}^3$ (Figure 3b).

Practice 4

The results for the short-term measurements of methacrylates during composite filling therapies in *Practice 4* are presented in Figure 4. The methacrylates MMA, EGDMA, and TEGDMA were detected during treatment. The concentrations for MMA varied from 70 to 400 $\mu\text{g}/\text{m}^3$ during composite filling therapies (Figure 4a). The exposure levels for EGDMA were between 8 and 13 $\mu\text{g}/\text{m}^3$ (Figure 4b). TEGDMA was detected with 42 and 45 $\mu\text{g}/\text{m}^3$, respectively (Figure 4c).

Discussion

In the past there has been a lack of knowledge about the exposure to volatile methacrylates in dental practices, but recently, a Swedish study presented results regarding the release of MMA and HEMA from bonding agents used in dental practices.⁵ The air levels of HEMA found in the Swedish study varied from 2 to 79 $\mu\text{g}/\text{m}^3$, which is in good agreement to our results with air levels between 7 and 45 $\mu\text{g}/\text{m}^3$. However, concentrations of MMA found in the Swedish study were much lower than our results. The maximum MMA concentrations in our study (up to 0.4 mg/m^3 , Figure 4a) were thirty times the concentrations found in Sweden. The reason for low MMA concentrations in the Swedish study is caused by focusing short-term measurements (1-18 min) on the use of bonding agents only, wherein MMA is more or less an impurity.⁵ Our study is focused on the exposure to volatile methacrylates in dental personnel during the whole filling therapy, taking into account every possible release of MMA. Beyond these data the results of our study show that dental personnel are exposed to various methacrylates.

MMA was detected in three dental practices in different concentrations (Figures 1a, 2a, 4a). Air levels of MMA occurred during the dental treatment at concentrations up to 0.4 mg/m^3 (Figure 4a) and is lower compared to concentration ranges in dental laboratories when liquid monomer is used directly containing more than 90 % MMA (5 mg/m^3).¹⁹ Thus, the results in this study show, that the exposure to MMA in dental personnel during filling therapies is at least 10 times lower than the MMA exposure in dental technicians during their work in dental laboratories.

The vapor pressure of HEMA is more than hundred times lower than the vapor pressure of MMA,⁵ nevertheless concentrations of HEMA up to 50 $\mu\text{g}/\text{m}^3$ were detected during composite filling therapies (Figures 1b, 2b, 3a). The sources for the release of HEMA are mainly the bonding agents used during filling therapies containing 20-40% HEMA.⁵ HEMA was detectable before three of the treatments started (Figures 1b, 3a), which might be caused by the release of HEMA during previous filling therapies. Nevertheless the detectable HEMA concentrations in dental practices were at about 10 times less than the MMA concentrations.

The methacrylate EGDMA was found in only one dental practice during three filling therapies with low concentrations at about 10 $\mu\text{g}/\text{m}^3$ (Figure 4b). The EGDMA concentration in the treatment room might not originate in the filling therapy but in a direct connection of the treatment room with a storage room for dental materials. A treatment independent EGDMA release can be explained as well by a constant EGDMA concentration between 11 and 13 $\mu\text{g}/\text{m}^3$ before, during and after one filling therapy (Figure 4b). Thus, the detected air levels of EGDMA are lower than the air levels of the other methacrylates found in dental practices.

The fourth methacrylate identified in the dental practices was the semi-volatile TEGDMA. TEGDMA, as HEMA, is a common ingredient of bonding agents⁵ and can be released during the treatment of the dentin surface with bonding agents. TEGDMA was detected in all dental practices in concentrations up to 45 $\mu\text{g}/\text{m}^3$ (Figures 1c, 2c, 3b, 4c). TEGDMA was not detectable before the treatment started and it is notable that TEGDMA was only detected once during a filling therapy in almost all of the 20 treatments, in which TEGDMA was detected. Obviously, the release of the semi-

volatile TEGDMA during filling therapies might cause only short-term exposures in dental personnel.

Nevertheless, the highest exposure levels of methacrylates in dental practices were achieved by MMA (Figure 5). The maximum concentrations of MMA were 10 times higher than HEMA and TEGDMA and 30 times higher than EGDMA, respectively. Maximum exposure levels at working environments without any harm to the workers are available only for MMA. The maximum allowable concentration (MAC value) for MMA in Germany is set to 210 mg/m³ and varies between 100 and 400 mg/m³ in other countries.^{3,30} The German MAC value of MMA allows short-term exposure levels (15 min) up to 420 mg/m³. The levels of MMA measured in this study with a maximum concentration of 0.4 mg/m³ are much lower than the MAC value and short-term threshold limit values. The low exposure levels of MMA in dental practices indicate that the toxicological risk for dental personnel is only of minor importance. No MAC values are available for HEMA, EGDMA, and TEGDMA. As the acute toxicity (LD₅₀, rat, oral) of MMA, HEMA, EGDMA, and TEGDMA are at about the same range (3-11 g/kg rat)^{28,30} but the detected highest exposure levels of HEMA, EGDMA, and TEGDMA are more than 2000 times lower than the MAC value of MMA, the toxicological risks of inhaled HEMA, EGDMA, and TEGDMA for dental personnel should be of minor importance as well.

Nevertheless, allergenic effects caused by methacrylates are well known and some methacrylates are strong sensitizers.^{2,14,16,21,29,32} Allergy tests with extensive methacrylate series on persons with suspected sensitivity on acrylic monomers indicated cross-reactivity between different acrylates.^{7,11} With measured concentrations of four different methacrylates in our study, the potential of allergenic cross-reactivity needs to

be taken into consideration, especially as allergenic reactions are not dependent on the occupational threshold limits. The appearance of contact dermatitis caused by methacrylates increased amongst dentists in recent years.¹³ The three methacrylates identified as responsible for most of occupational contact allergies in dentistry were HEMA, EGDMA, and TEGDMA.⁴ Although most of the studies mentioned above represent contact allergenic reactions, they substitute the scarce information about allergic reaction of dental personnel caused by inhaled methacrylates. Piirila *et al.* reported occupational respiratory diseases, including asthma, due to methacrylates in dental personnel in Finland.²³ Moreover, occupational respiratory hypersensitivity amongst dental personnel increased rapidly since the 1990s and seems to be connected with the replacement of amalgam fillings with acrylic-based polymers.²²

Therefore the data presented show that dental personnel are exposed to various volatile methacrylates such as MMA, HEMA, EGDMA, and TEGDMA during dental filling therapy.

Clinical Relevance

Our results demonstrate that the exposure levels of methacrylates in the air of dental practices during filling therapies are lower than the threshold limit values. However, an observable increase of allergic respiratory diseases amongst dental personnel due to methacrylates exposure is not dependent on these toxicological threshold limit values. To minimize any possible health risks (e.g. allergic respiratory diseases such as asthma) for dental personnel a reduction of the air levels of methacrylates in dental practices should always be attempted.

Acknowledgements

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Figures
Figure 1a

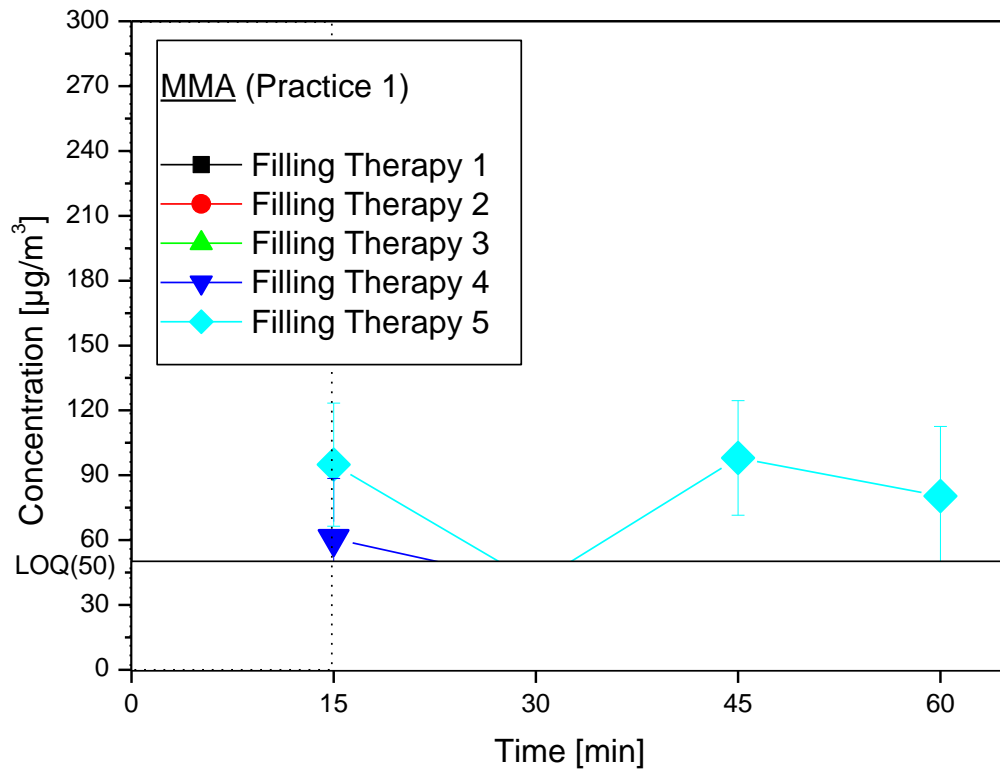


Figure 1b

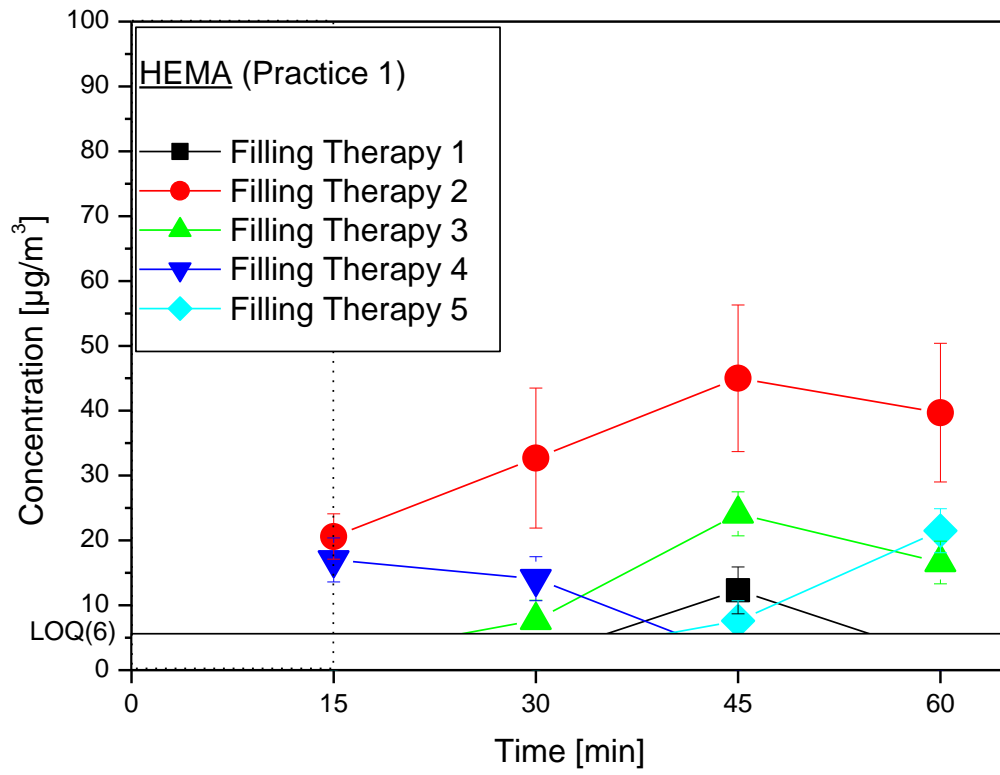


Figure 1c

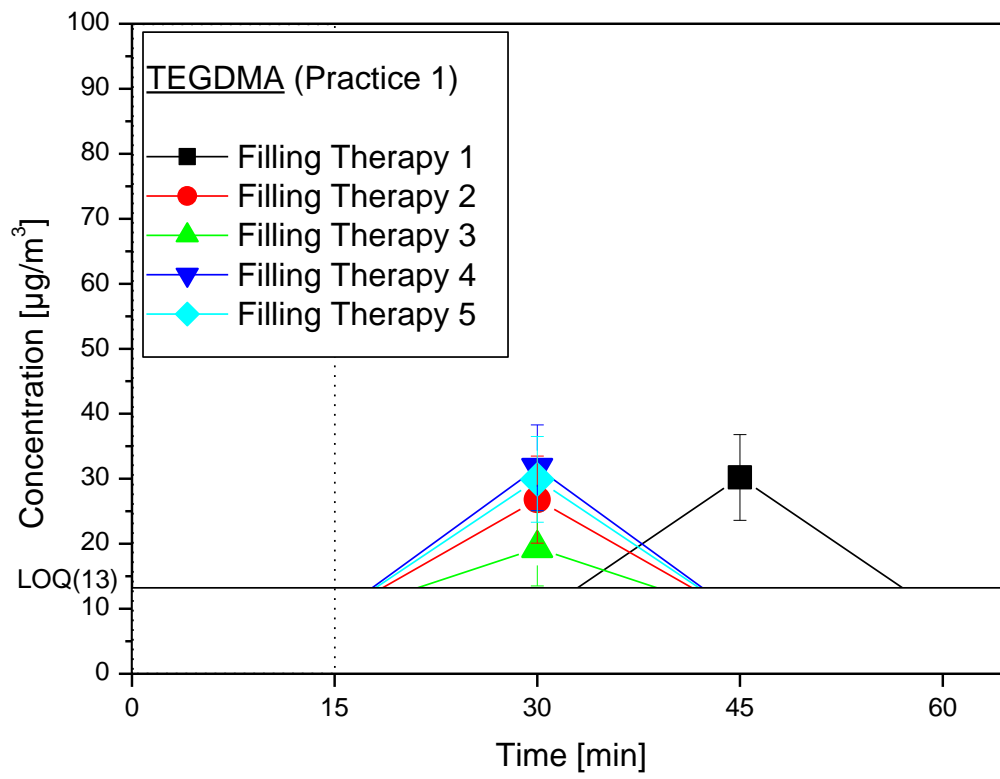


Figure 2a

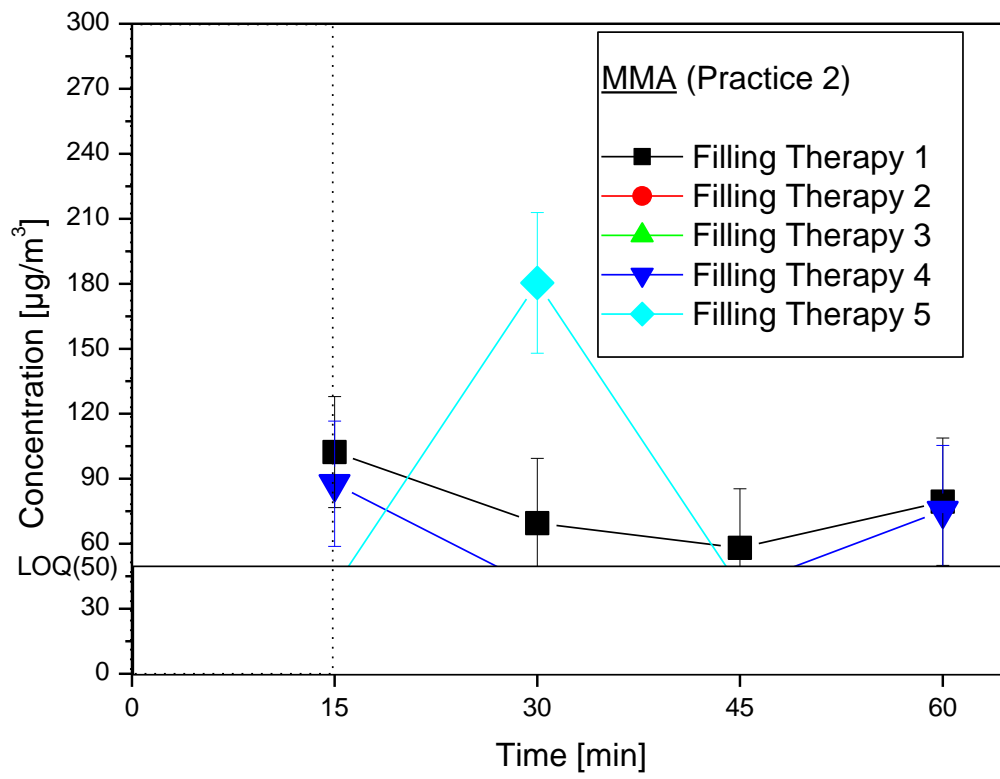


Figure 2b

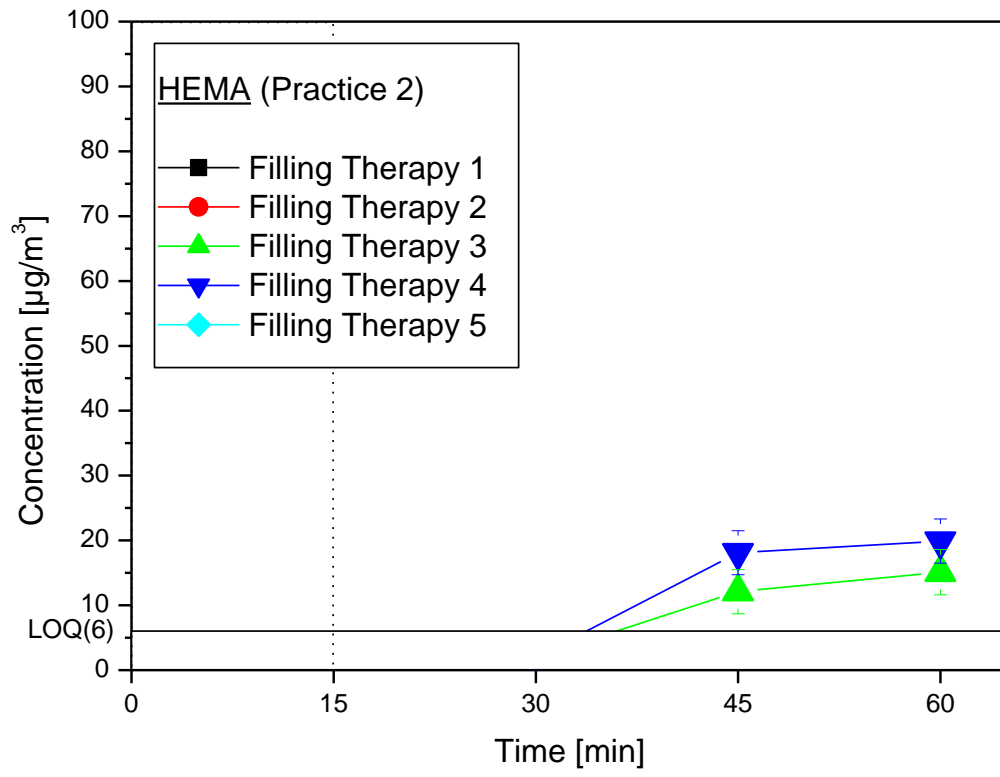


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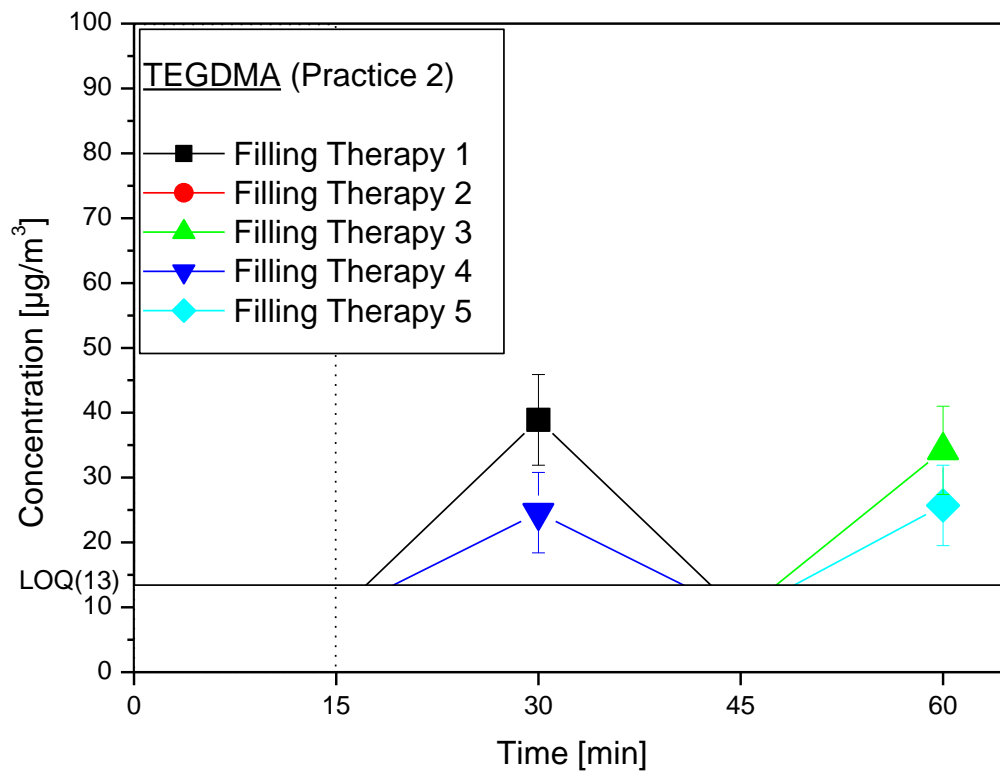


Figure 3a

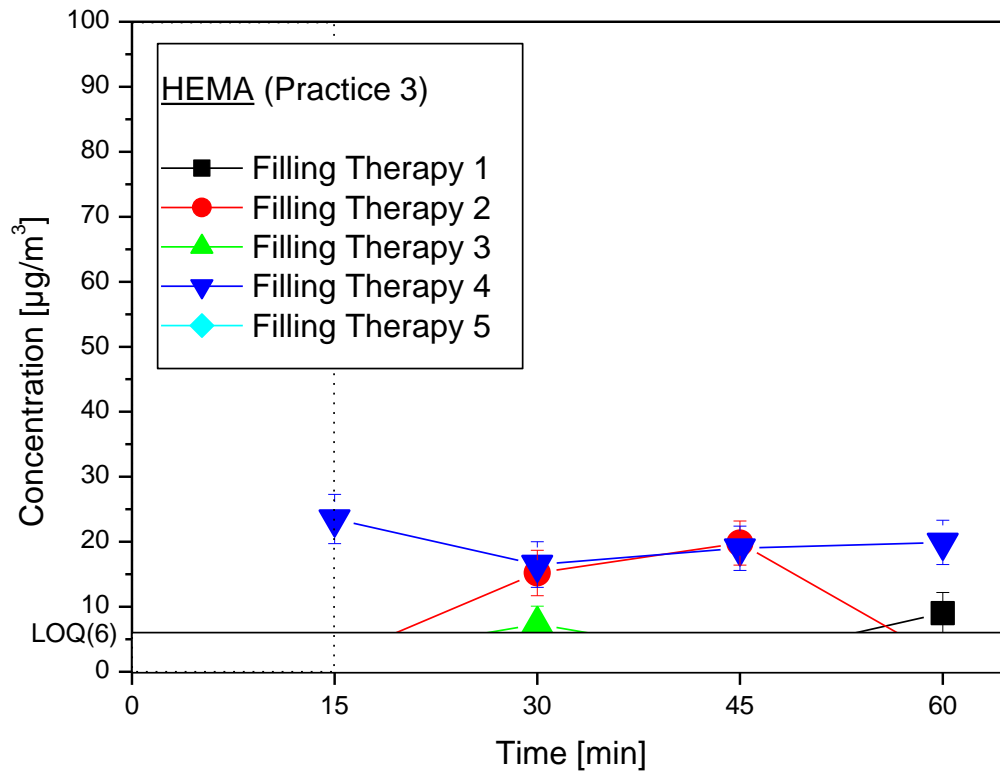


Figure 3b

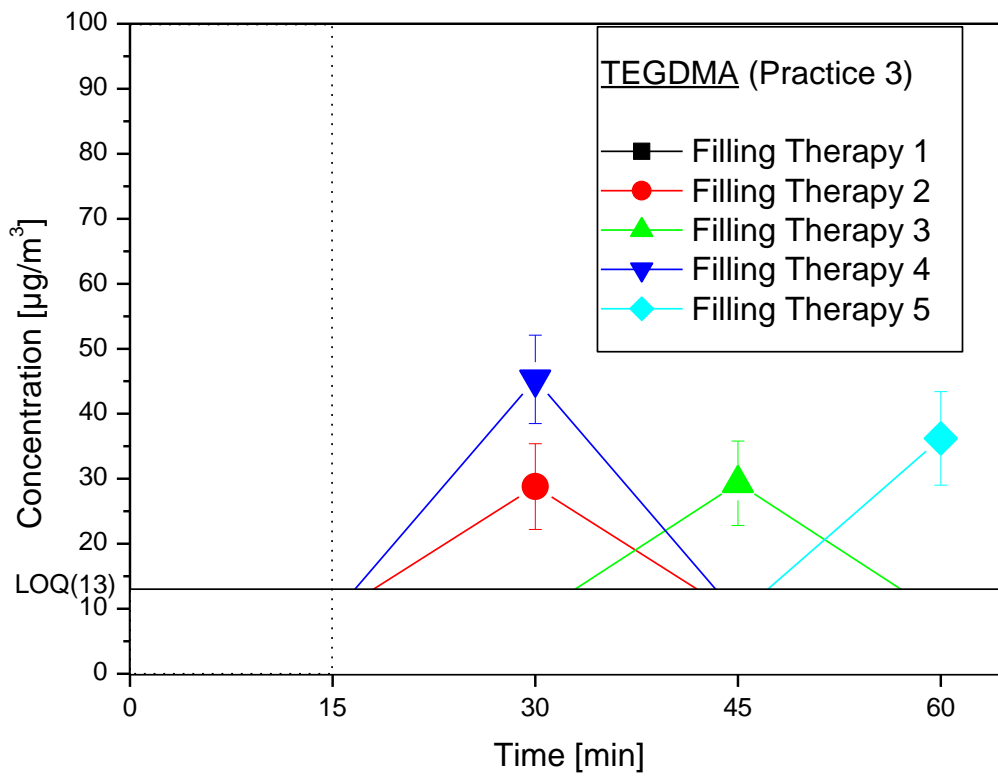


Figure 4a

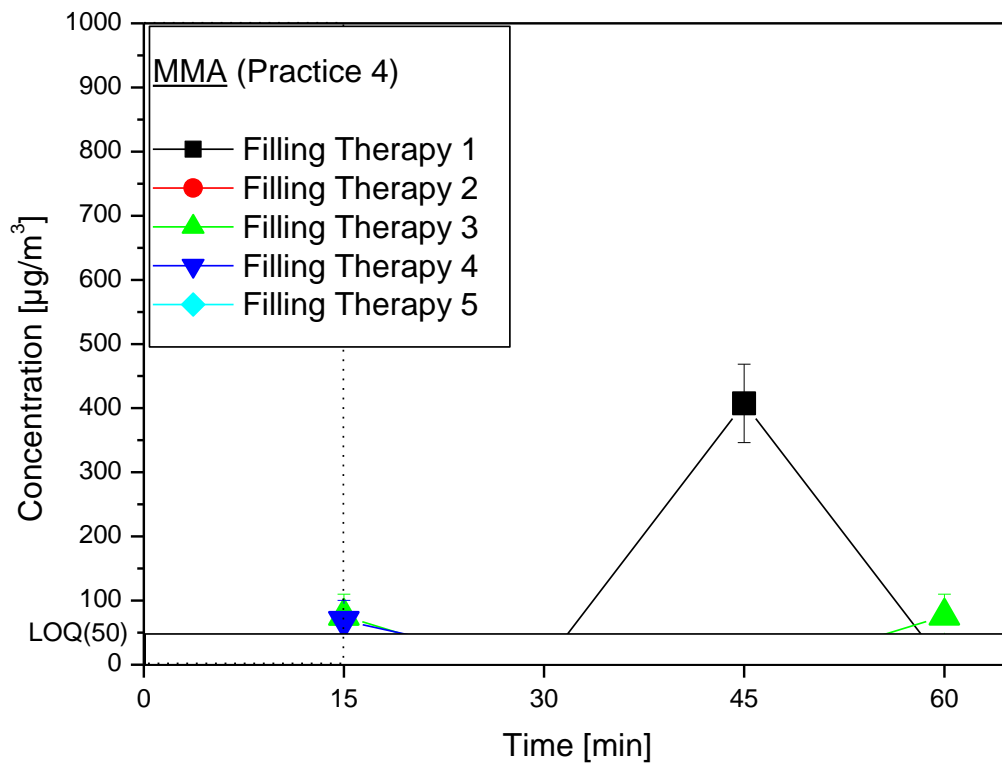


Figure 4b

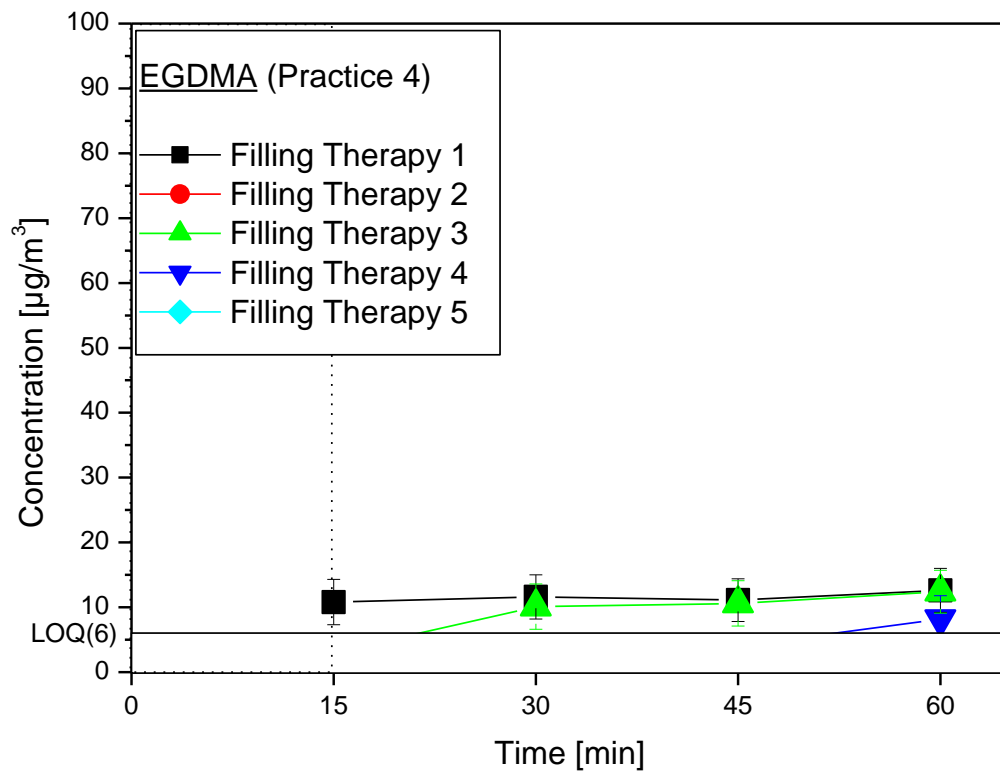


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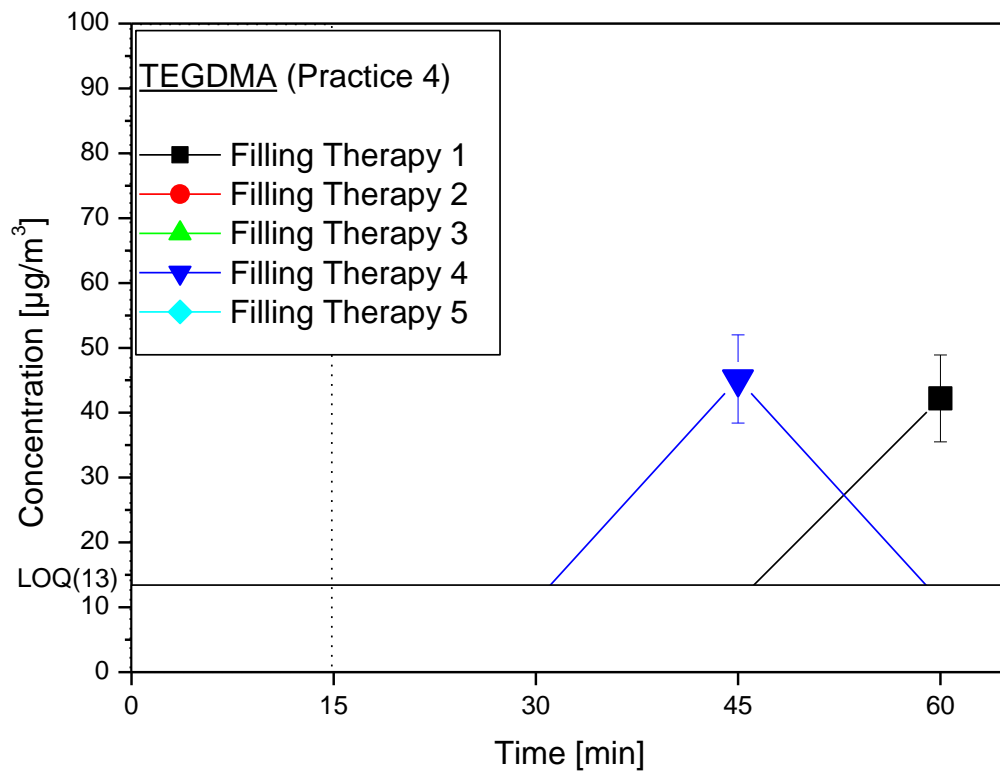
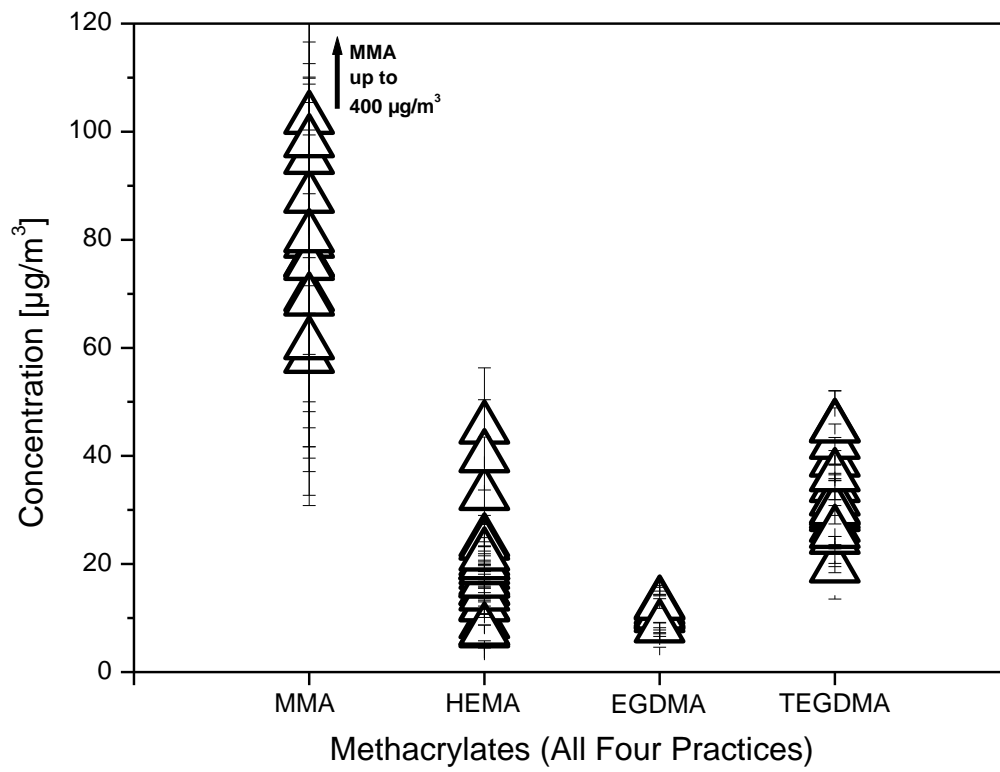


Figure 5



Legends

Figure 1: Exposure to methacrylates at 20 short-term measurements (15 min) in Practice 1 (air sampling before and during five dental therapies in between one hour with four sampling fibers, start of sampling at 0 min, start of dental treatment at 15 min). The error bars represent the 95 % confidence intervals (95 % CI), LOQ is the limit of quantification. (a) Exposure to MMA, (b) Exposure to HEMA, (c) Exposure to TEGDMA.

Figure 2: Exposure to methacrylates at 20 short-term measurements (15 min) in Practice 2 (air sampling before and during five dental therapies in between one hour with four sampling fibers, start of sampling at 0 min, start of dental treatment at 15 min). The error bars represent the 95 % confidence intervals (95 % CI), LOQ is the limit of quantification. (a) Exposure to MMA, (b) Exposure to HEMA, (c) Exposure to TEGDMA.

Figure 3: Exposure to methacrylates at 20 short-term measurements (15 min) in Practice 3 (air sampling before and during five dental therapies in between one hour with four sampling fibers, start of sampling at 0 min, start of dental treatment at 15 min). The error bars represent the 95 % confidence intervals (95 % CI), LOQ is the limit of quantification. (a) Exposure to HEMA, (b) Exposure to TEGDMA.

Figure 4: Exposure to methacrylates at 20 short-term measurements (15 min) in Practice 4 (air sampling before and during five dental therapies in between one hour with four

sampling fibers, start of sampling at 0 min, start of dental treatment at 15 min). The error bars represent the 95 % confidence intervals (95 % CI), LOQ is the limit of quantification. (a) Exposure to MMA, (b) Exposure to EGDMA, (c) Exposure to TEGDMA.

Figure 5: Exposure levels of MMA, HEMA, EGDMA, and TEGDMA at 80 short-term measurements (15 min) in four different Practices. MMA exposure levels exceeding $120 \mu\text{g}/\text{m}^3$ ($0.2 \text{ mg}/\text{m}^3$ and $0.4 \text{ mg}/\text{m}^3$) are not shown in the figure. The error bars represent the 95 % confidence intervals (95 % CI).