



Spatial variability in the ambient concentrations of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans across the Middle East

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ABSTRACT

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/F) were monitored at 12 sites across four countries (Kuwait, Turkey, Lebanon and, Oman) using validated passive air samplers between January 2018 and December 2018 in an attempt to set up a monitoring network for persistent organic pollutants (POPs) across the Middle East. The highest concentrations of Σ PCDD/F were detected in Oman reflecting the proximity of the site to a steel melting company at Al-Rassil Industrial area (557 fg I-TEQ m⁻³) and a hospital waste incinerator at Al-Multaga (489 fg I-TEQ m⁻³). Elevated levels were also measured at all monitoring sites across Kuwait with average concentrations ranging from ~150 fg I-TEQ m⁻³ at the Shuwaikh site to ~250 fg I-TEQ m⁻³ at the Salmiya and Abdali sites. Levels of Σ PCDD/F at all other stations in the region were generally lower than 35 fg I-TEQ m⁻³. The levels of 1,2,3,4,7,8-HxCDF were frequently a factor of two higher compared to 1,2,3,6,7,8-HxCDF suggesting that an additional source exists which involves the use of elemental chlorine. This study has provided the first reliable measurements of the atmospheric concentrations of PCDD/F in the Middle East providing a baseline for assessing time trends in PCDD/F in the air, one of the core matrices for the effectiveness evaluation under the Stockholm Convention on POPs.

1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are two examples of unintentionally produced persistent organic pollutants (POPs) that are characterized by their persistence, bioaccumulative potential, susceptibility to long-range transport, and adverse health effects (Muir and de Wit, 2010; Weber et al., 2011). PCDDs and PCDFs originate primarily from industrial activities such as metal smelting, incineration of medical waste, processing paper pulp with elemental chlorine, and backyard burning of household waste containing chlorinated materials such as plastics, in particular, those made from polyvinyl chlorides (PVCs). The production of

chlorinated aromatics such pentachlorophenol (PCP), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), pentachloronitrobenzene, 2,4-D, and other organochlorines results in PCDD/F production as byproducts (Holt et al., 2010; UNEP, 2013b). This source is thought, historically, to be the largest source of environmental PCDD/F pollution (Camenzuli et al., 2015; Weber et al., 2008). Natural PCDD/F sources include forest fires, which are primarily a redistribution of adsorbed PCDD/F (Prange et al., 2003). The combination of their resistance to metabolism degradation and lipophilicity leads to their bioaccumulation and transport through food chains (Daley et al., 2011; Ikemoto et al., 2008; Kelly et al., 2007). These concerns led to the Stockholm Convention (SC) on POPs, international treaty aiming to

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reduce and ban these chemicals.

The Stockholm Convention's Article 16 mandates that the signatory countries evaluate the effectiveness/success of the regulations implemented by the Convention to eliminate or reduce POP releases into the environment on a regular basis. For this purpose, the Global Monitoring Plan (GMP) was formed by the Convention to gather comparable, harmonised, and reliable data on POP levels in key environmental matrices such as air, breast milk/blood, and water.

About 95% of all PCDD/F are currently emitted into the atmosphere and the residual 5% is released to the aquatic environment, or the soil (Vikelsøe et al., 2006).

Once released, PCDD/F are mainly transported through the atmosphere and deposited in terrestrial and aquatic compartments, eventually reaching remote locations with no POP sources that can contaminate soil and sediments. Human exposure to dioxin-like chemicals is often associated with ingesting contaminated foods. In Germany, the primary source of PCDD/F contamination of food of animal origin is soils contaminated from past PCDD/F releases (Weber et al., 2018). Air is one of the two core media selected by the SC for monitoring POPs in the environment (UNEP, 2007, 2013a) and it is the major route of long-range transport through the environment (UNEP, 2009, 2013a).

Since POPs are subject to long-range transport (LRT) from locations where they are produced to regions where they have never been used, reliable environmental data from every part of the world is required to properly understand the key processes driving worldwide dispersion of these chemicals (Meng et al., 2011; Scheringer et al., 2004). At present, several monitoring programs are being carried out at national, regional, and global basis such as the Global Monitoring Plan (GMP) for SC, The Arctic Monitoring and Assessment Programme (AMAP), the Northern Contaminants Programme (NCP), Monitoring NETWORK (MONET), the European Monitoring and Evaluation Program (EMEP), the Working Group on Effects (WGE) for Convention on Long-range Transboundary Air Pollution (CLRTAP) or Global Aquatic Passive Sampling (AQUA-GAPS).

Although environmental data on POPs are available for some areas of the world, there is a scarcity of scientific data on these chemicals throughout the Middle East, Africa, and Asia. Data is needed from all regions of the world to evaluate the efficiency of international initiatives to reduce their release into the environment. In this study, passive samplers were deployed at 12 sites across four Middle Eastern countries (Kuwait, Turkey, Lebanon and Oman) between January and December 2018 to redress this balance and to provide comparable monitoring data across the Middle East. These samples were retrieved every three months and the PUF disks were kept in amber glass jars and stored at 4 °C until they were shipped to Kuwait for analysis. These deployment approaches were designed to analyze seasonal variability in ambient air concentrations of POPs as well as to obtain information on the spatial distribution of these chemicals throughout the Middle East. A secondary objective of this study is to set up an air monitoring network in the Middle East for POPs to monitor trends in their ambient air concentrations. POPs monitoring programs are crucial to evaluate the effectiveness of the measures adopted by the SC on reducing the emission of these pollutants to the environment. The deployment of passive samplers (PAS) is a simple and cost-effective alternative to active air samplers (Harner et al., 2004; Jaward et al., 2004a; Pozo et al., 2004; Shoeib and Harner, 2002). The GAPS monitoring network, for example, was initiated in 2005 across 55 global sites to support the GMP is solely based on passive sampling. Additionally, the passive sampling technique is applied along with active sampling technique in other POPs monitoring networks such as MONET and NCP.

Spatial mapping of POP concentrations on any scale (local, regional, or global) might be employed as a valuable tool to identify the sources, localised 'hot spots', and also the understanding of transport processes.

Therefore, in many places, including developing countries where POPs research is still in its infancy, PAS are the sampler of choice. It was determined from earlier investigations that the samplers employed in

the current study produce data that are compatible with those obtained using high-volume air samplers (Abad et al., 2022; Bartkow et al., 2004; Jaward et al., 2004b; Rauer et al., 2018; Schuster et al., 2015a; Shoeib and Harner, 2002; Wania and Shunthirasingham, 2020).

As it is stated by Schuster et al. (2021), enhancing data comparability from various POPs monitoring programs should receive major consideration in order to minimize regional information gaps and, in this context, the current study aims to serve as a key research activity to initiate regular monitoring studies in the Middle East region.

2. Materials and methods

2.1. Chemicals and reagents

The calibration set for the analysis of PCDD/F (EDF-4947) was purchased from Cambridge Isotope Laboratories (CIL, MA, USA). The sampling standard (EN1948SS), the extraction standard (EN-1948ES: 2,3,7,8-Tetrachloro [¹³C₁₂]dibenzo-p-dioxin; 2,3,7,8-Tetrachloro [¹³C₁₂]dibenzofuran; 1,2,3,7,8-Pentachloro [¹³C₁₂]dibenzo-p-dioxin; 2,3,4,7,8-Pentachloro [¹³C₁₂]dibenzofuran; 1,2,3,4,7,8-Hexachloro [¹³C₁₂]dibenzo-p-dioxin; 1,2,3,6,7,8-Hexachloro [¹³C₁₂]dibenzo-p-dioxin; 1,2,3,4,7,8-Hexachloro [¹³C₁₂]dibenzofuran; 1,2,3,6,7,8-Hexachloro [¹³C₁₂]dibenzofuran; 2,3,4,6,7,8-Hexachloro [¹³C₁₂]dibenzofuran; 1,2,3,4,6,7,8-Heptachloro [¹³C₁₂]dibenzofuran; Octachloro [¹³C₁₂]dibenzo-p-dioxin; Octachloro [¹³C₁₂]dibenzofuran), and recovery standard (EN-1948IS) (1,2,3,4-Tetrachloro [¹³C₁₂]dibenzo-p-dioxin and 1,2,3,7,8,9-Hexachloro [¹³C₁₂]dibenzo-p-dioxin) were acquired from Wellington Laboratories (ON, Canada). PUF plugs and passive samplers were obtained from Tisch Environmental (OH, USA). Clean-up columns (multilayer silica, basic alumina and PX-21 carbon) were purchased from FMS, Inc (MA, USA). Pesticide grade dichloromethane, hexane, ethyl acetate and toluene acquired from Fisher Scientific (UK), were used for the extraction and clean-up of the target analytes. The preparation of the spiking solutions was carried out in nonane and tetradecane obtained from Sigma Aldrich and Carlo Erba, respectively.

2.2. Sampling locations and sampling

Initially, it was planned to deploy passive samplers in Bahrain, Oman, Qatar, United Arab Emirates, Kuwait, Lebanon, Yemen, Syria, Saudi Arabia, Turkey and, Jordan. However, due to ongoing armed conflicts in the region, the sampling was limited to certain countries where sampling could be completed safely.

The samplers were therefore placed at locations across the Middle East representing different regional characteristics such as agricultural, industrial, rural, urban, and backdrop along a northwest-southeast transect. Additionally, political instability and logistics-related complications led to sampling deficiencies resulting in the inability to deploy samplers at all sites for all sampling campaigns. Briefly, sampling started in January 2018 in Turkey and Kuwait, while it started in February or in March in other countries. Sampling ended at the end of 2018 in Turkey, while it ended in late March/early April in 2019 in other countries. Hence, four sampling campaigns were conducted in Turkey, Oman and Lebanon, while five sampling campaigns were carried out in Kuwait (Supporting Information Table S1). The selected sites were as follows: two sites in Turkey (Uludag and Turkish Scientific Research Council Bursa Test and Analysis Laboratory (BUTAL)); two sites in Oman (Al Rassil and Al Multaga), three sites in Lebanon (Campus of American University of Beirut (AUB), Ashrafieh and Qartaba) and, five sites in Kuwait (Salmiya, Abdali, Ahmadi, Sulaibiya and Shuwaikh). Details on characteristics of the sampling locations are given in Supporting Information Table S1. A sampling kit comprising of dome shaped passive sampling housing units, pre-cleaned polyurethane foam (PUF) disks (two PUF disks for concurrent sampling at each sampling site), acetone-dichloromethane rinsed collection glass jars, and gloves were dispatched

in December 2017 to all participants that volunteered to participate in the study. The package also included a standard operating procedure (SOP), which was based on the protocol established by the Global Atmospheric Passive Sampling (GAPS) Network run by Environment and Climate Change Canada, for deployment and retrieval of samplers. The samplers were retrieved approximately every three months (see Supporting Information Table S1 for details). At the end of each sampling period, the PUF disks were stored at $-20\text{ }^{\circ}\text{C}$ until they were shipped to the "Stockholm Convention Sub-Regional Centre for POPs" laboratory facilities in Kuwait (SCRC Kuwait) for analysis.

Dome-shaped passive sampling devices (Shoeib and Harner, 2002), deployed at $\sim 2\text{--}2.5\text{ m}$ from the ground in unobstructed airflow were used to collect air samples in the current study. Flame retardant free certified as PUF disks were purchased from Tisch Environmental (OH, USA) and were cleaned using a series of solvent mixtures in a giant Soxhlet (1.5 L, 46 inches Soxhlet apparatus, fritted top: 103/60; fritted bottom: 45/50, Ace Glass Incorporated, NJ, USA), each solvent extraction over a 24 h period; acetone-dichloromethane; acetone-toluene and acetone-methanol). To avoid contamination before deployment, the pre-extracted PUF disks were dried in a clean desiccator under vacuum and stored in solvent-rinsed glass jars. Although this study is part of a broader project related to ambient air concentrations of legacy POPs (Gevao et al., 2022) only PCDD/F concentrations are reported in the current manuscript.

2.3. Extraction and clean-up

Hexane-dichloromethane (1:1) mixture was used for extraction of target chemicals from two PUF disk passive samplers collected simultaneously at each location. Extraction was carried out on an accelerated solvent extractor (ASE-350, Dionex, CA, USA) equipped with 33 ml stainless-steel extraction cells. Samples were spiked with a total of 400 pg of the $^{13}\text{C}_{12}$ -labelled extraction standard (EN-1948ES). In accordance with US-EPA Method 3545a (USEPA, 2007), the cell was sealed and placed in the extractor carousel 2 h later to begin the extraction process on the following conditions: temperature $100\text{ }^{\circ}\text{C}$, pressure 1500 psi, static time 5 min, 1 extraction cycle.

Clean-up and fractionation of extracts was conducted on an automated multi-column sample cleanup system (Power Prep™; FMS, Inc, MA, USA) while instrumental analysis was carried out on an AutoSpec premier high-resolution mass spectrometer (Waters, Manchester, UK) coupled with an Agilent 7890 gas chromatograph equipped with 2 split/splitless inlets and 2 fused silica columns. Chromatographic separation of PCDD/F was achieved on a DB-5 MS (60 m \times 0.25 mm i. d. X 0.25 μm film thickness; J&W Scientific, CA, USA). Target analytes were quantified by the isotope dilution method; full details of the cleanup and the final determination method have been previously reported (Abad et al., 2007; Martinez-Guijarro et al., 2017).

2.4. Deriving air concentrations and air volume estimation

The chemical amount accumulated in PUF disks (ng/sampler) divided by the effective air volume to calculate the concentrations of PCDD/F in the air (V_{AIR} , m^3) (Shoeib and Harner, 2002). Derivation of air volumes based on the loss of deuration compounds is a widely followed method in literature (Gouin et al., 2005; Pozo et al., 2004) however, estimation of air concentrations using an assumed air sampling rate (Harner et al., 2002; Jaward et al., 2004a) or employment of a model that uses local weather data are also methods available in the published literature (Jahnke et al., 2022; Llanos et al., 2022; Schuster et al., 2021). In the current study, effective sampling volume (R , m^3/day) was derived from the model (Herkert, 2016) to estimate the concentrations of PCDD/F in ambient air. For this purpose, ambient air temperature data for sampling sites were obtained from the National Oceanic and Atmospheric Administration (available at <https://www.ncdc.noaa.gov/cdo-web/>). The model-derived average R -value ranged

between 2.24 and 4.63 m^3/day for sampling sites. The Excel spreadsheet created for the GAPS Network (Harner, 2017) was employed by using the sampling site-specific R -value, ambient air temperature data and total deployment duration (days) to calculate the total sampled air volume for each target compound. Compounds-specific total sampled air volume ranges between 273 and 360 m^3 over the three-month deployment period and this chemical-specific total air was used to calculate air concentrations of the target chemicals.

2.5. Quality assurance/Quality control (QA/QC)

The proper performance of the analytical procedure was assured by spiking each sample with labelled compounds, analyzing the procedural blanks (one blank PUF with each set of collected samples), and participating in international inter-calibration exercises such as INTERCIND and UNEP. No blank concentration correction was performed on the data obtained. Recovery rates of individual labelled compounds are given in Supporting Information Table S2 and it ranged between 30% and 136%, however, congeners with recovery rates less than 50% did not contribute to more than 10% of the total I-TEQ in the relevant sample. Overall, in all samples, the average recovery rates of the extraction standards ranged from $63 \pm 19\%$ to $106 \pm 19\%$ meeting the requirements of the reference analytical methods for these analytes such as EN1948 European standard (EU, 2006) and EPA method 1613 (US-EPA, 1994). Additionally, each sequence run included a detailed evaluation of the GC column's performance as well as the sensitivity and resolution power (maintained at 10,000, 10% valley) of the HRGC/HRMS system.

The analyte concentration that generated a signal higher than three times the standard deviation (SD) of the noise level was defined as the instrument detection limit (IDL). In case an analyte was observed in a laboratory blank that was processed alongside a batch of samples, the average concentration of the blanks was deducted from the concentration detected in the sample. The method detection limit (MDL) was defined as the average concentration of target analyte in field blanks plus 3 X standard deviation (3SD). IDL was accepted to be equal to MDL in cases when the targeted analytes were not present in the blanks. To convert MDL values to I-TEQ fg/m^3 , a final sample volume of 50 μL and an average air volume of 350 m^3 were used. IDL ($\text{fg}/\mu\text{L}$) and MDL (I-TEQ fg/m^3) values are given in Supporting Information Table S3.

3. Results and discussion

3.1. Atmospheric concentrations and spatial variability of PCDD/F

The $\Sigma 2,3,7,8$ chlorinated PCDD/F concentrations measured at each site are summarised in Fig. 1, whereas the congener-specific concentrations for each sampling period are given in Supporting Information Tables S4, S5, S6 and S7 for samples collected across the Middle East and at the four sites in Kuwait, respectively. The two highest $\Sigma\text{PCDD/F}$ concentrations were measured in the vicinity of a secondary steel smelting company at Al-Rassil Industrial area (557 fg I-TEQ m^{-3}) and a hospital incinerator at Al-Multaga (489 fg I-TEQ m^{-3}) in Oman during the first sampling campaign (March–June 2018) indicating that these point sources impacted these sampling locations. The lowest concentrations were measured at the sampling site Lebanon-Ashrafieh (10.8 fg I-TEQ m^{-3}). The concentrations measured at all three sites in Lebanon generally ranged from 21 to 33 fg I-TEQ m^{-3} and between 19 and 31 fg I-TEQ m^{-3} at both sites in Turkey. Elevated levels were observed across Kuwait with average concentrations measured ranging from $\sim 150\text{ fg I-TEQ m}^{-3}$ at Shuwaikh to 250 fg I-TEQ m^{-3} at the Salmiya and Abdali (Fig. 2).

Consecutive measurements were carried out at several sites in Kuwait (Fig. 2), Turkey (Fig. 3), Lebanon (Fig. 4) and Oman (Fig. 5). No substantial differences were observed except in Oman where noticeable variations in the concentrations were measured over the sampling

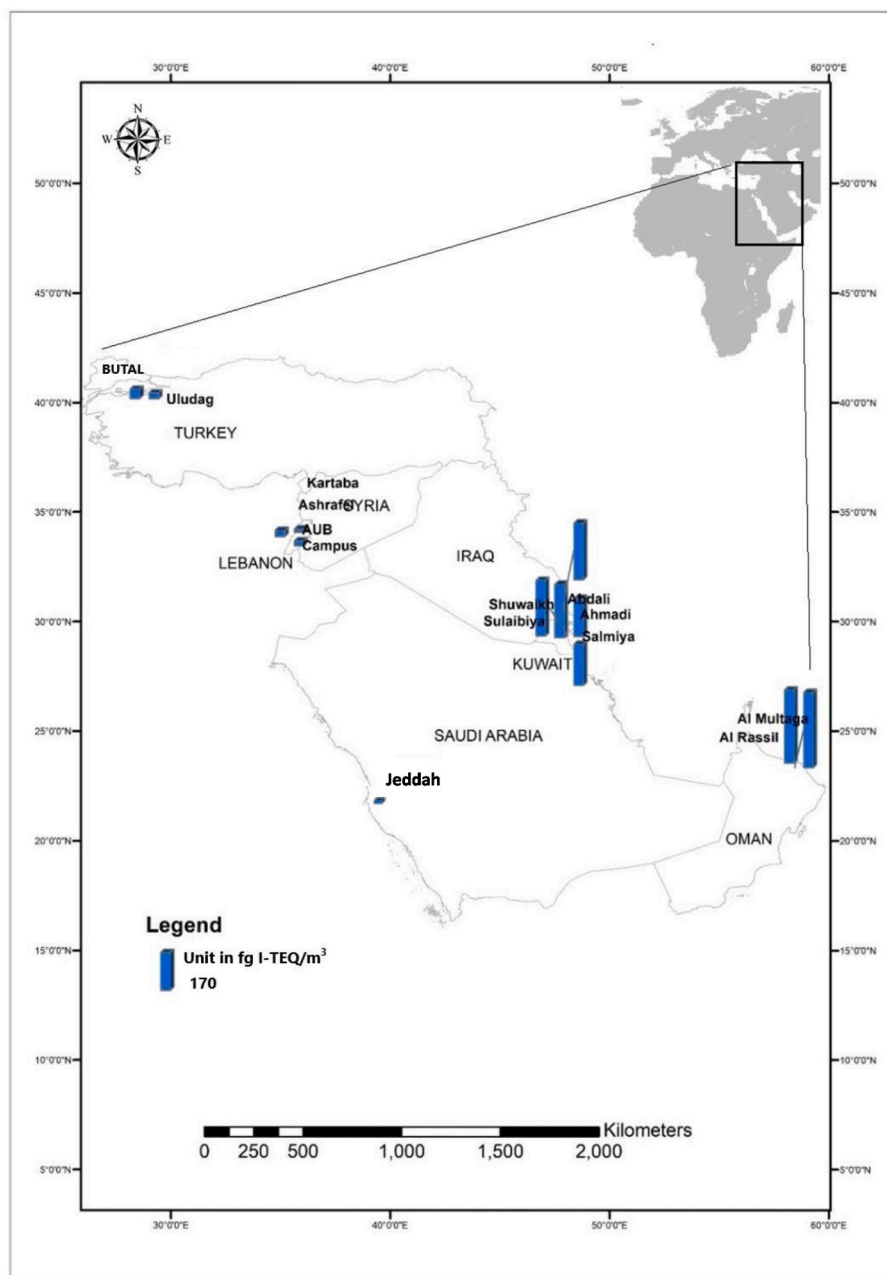


Fig. 1. Spatial variations in the air concentrations of Σ PCDD/F (fg I-TEQ/m³) air samples from four Middle Eastern countries between January 2018 and December 2018.

period (Fig. 5). The PCDD/F concentrations measured at Al Rassil ranged from 92 to 557 fg I-TEQ/m³ (mean: 291; median: 258) and from 54 to 489 fg I-TEQ/m³ at Al Multaga (mean: 194; median: 116). This might indicate reduced releases from the two mentioned point sources but no particular assessment of the facilities was conducted. Also, secondary metal smelters can have a variation in PCDD/F emission depending on the scrap intake (Wu et al., 2020; Yang et al., 2020).

Although there is no statistically significant correlation with the mean temperatures (Fig. 2), the mean summer concentrations exceeded the mean winter concentrations at all the urban sites in Kuwait where measurements have been carried out over five consecutive campaigns (Fig. 6). This differs from the time trend in more northern countries where PCDD/F levels in summer are lower than in winter (Coutinho et al., 2007; Hovmand et al., 2007). This observation suggests that the atmospheric variations at the urban sites in Kuwait City may be driven by factors that slightly somewhat increase emissions in summer. The

increased power generation for air-conditioning during the very hot summers in Kuwait might have higher PCDD/F emissions from power stations located across the country (Fiedler, 2007; Wielgosiński, 2011). However, the opposite trend was observed at the Abdali site with moderately elevated high wintertime concentrations. The high Σ PCDD/F concentrations at the “remote” site at Abdali throughout the year of 250 (141–342) fg I-TEQ/m³ [mean (range)] may be due to its proximity to oil operations in the north of Kuwait which generates PCDD/F from flaring and from catalytic cracking and regeneration of the catalyst (UNEP 2013a,b; Government of Kuwait 2020). This sampling point is therefore not related to open waste burning but to an industrial point source, therefore, the area would depend on the oil production volumes. High PCDD/F concentrations in ambient air samples were previously reported at Umm-Al-Aish oil field, Kuwait (Martinez-Guijarro et al., 2017).

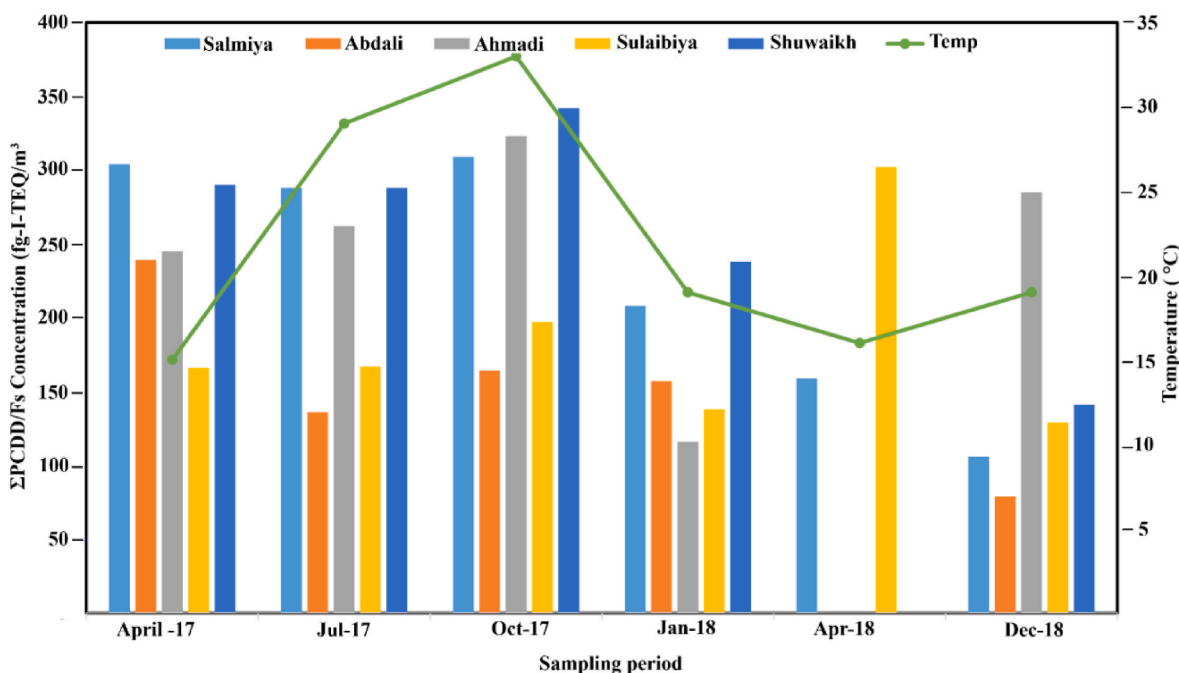


Fig. 2. Seasonal Variations in Concentrations of Polychlorinated Dibenzo-*p*-Dioxins and Dibenzofurans at five Locations across Kuwait between April 2017 and December 2018.

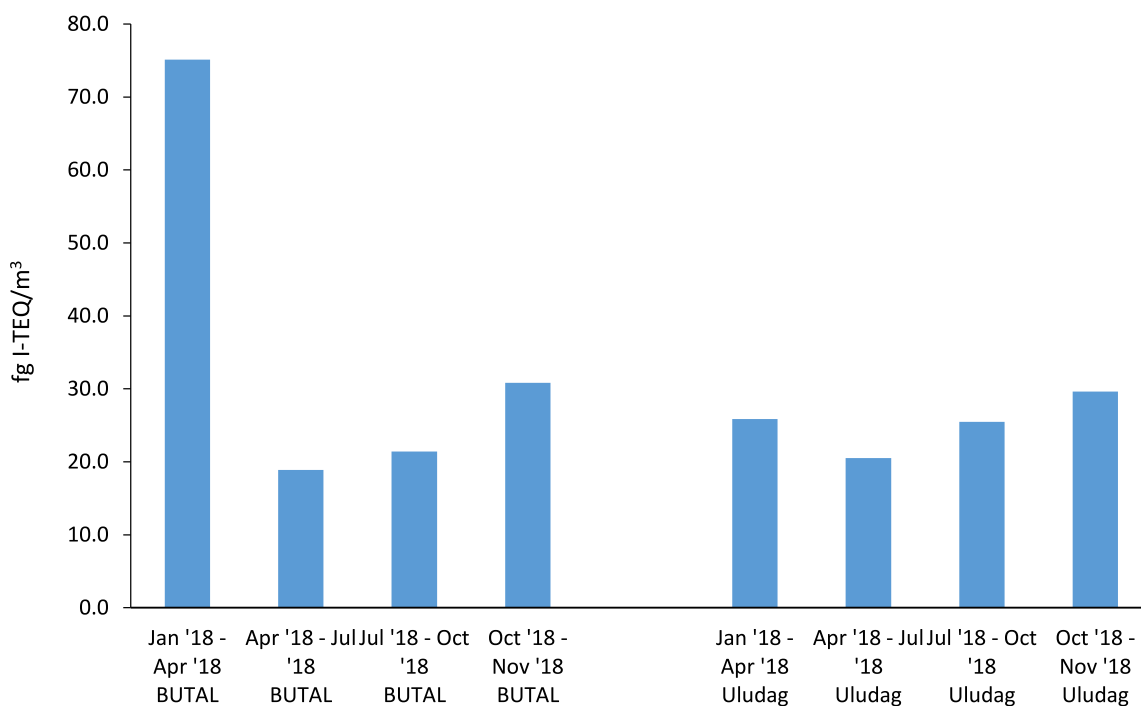


Fig. 3. Seasonal variations in concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans at two locations in Turkey between January 2018 and November 2018.

3.2. Comparison to published PCDD/F levels

The TEQ values measured in this study ranged between 4 and 30 fg I-TEQ m⁻³ except for Kuwait and Oman where elevated concentrations of 217 and 242 fg I-TEQ m⁻³ respectively were measured. Two separate studies carried out in the Group of Latin America and the Caribbean (GRULAC) region using the same type of passive samplers as the current study reported values ranging between 3.6 and 25 fg I-TEQ m⁻³

(Schuster et al., 2015b), and 34 fg I-TEQ m⁻³ in Sao Paulo, Brazil, and 5.3 fg I-TEQ m⁻³ at a site in Quito, Ecuador (Bogdal et al., 2013). The study by Bogdal et al. (2013) also reported TEQ values at some sites in Africa, and Pacific Island states of a similar order of magnitude to those reported for Lebanon and Turkey in this study. The elevated concentrations in Oman and Kuwait are identical to those reported using passive samplers in several cities in Seoul-South Korea (209–810 fg I-TEQ m⁻³) (Yoonki et al., 2014); in Bou-Ismaïl- Algeria (210–350 fg I-TEQ

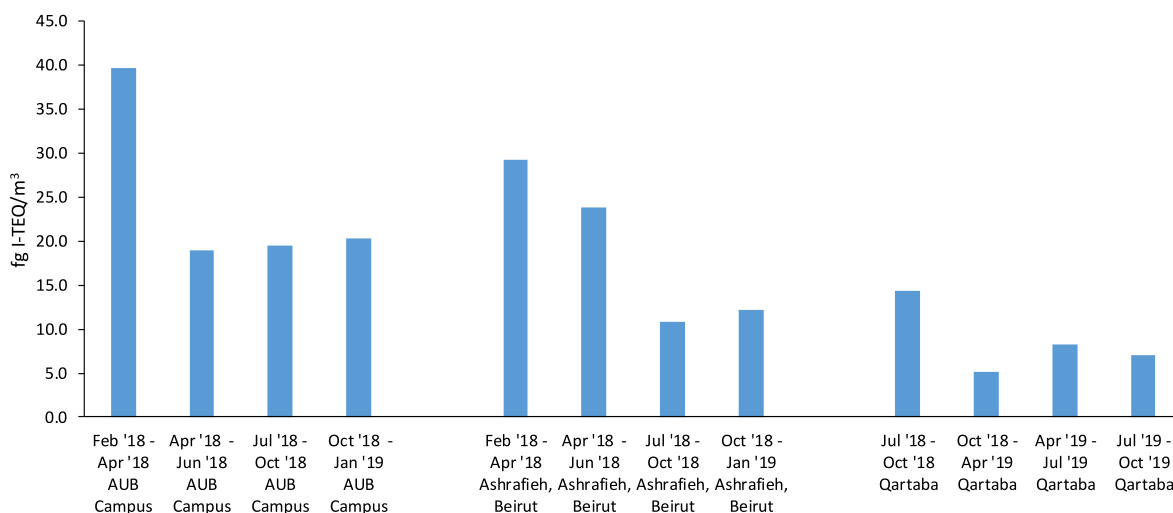


Fig. 4. Seasonal variations in concentrations of polychlorinated dibenzo-p-dioxins and dibenzofurans at three locations in Lebanon between February 2018 and October 2019.

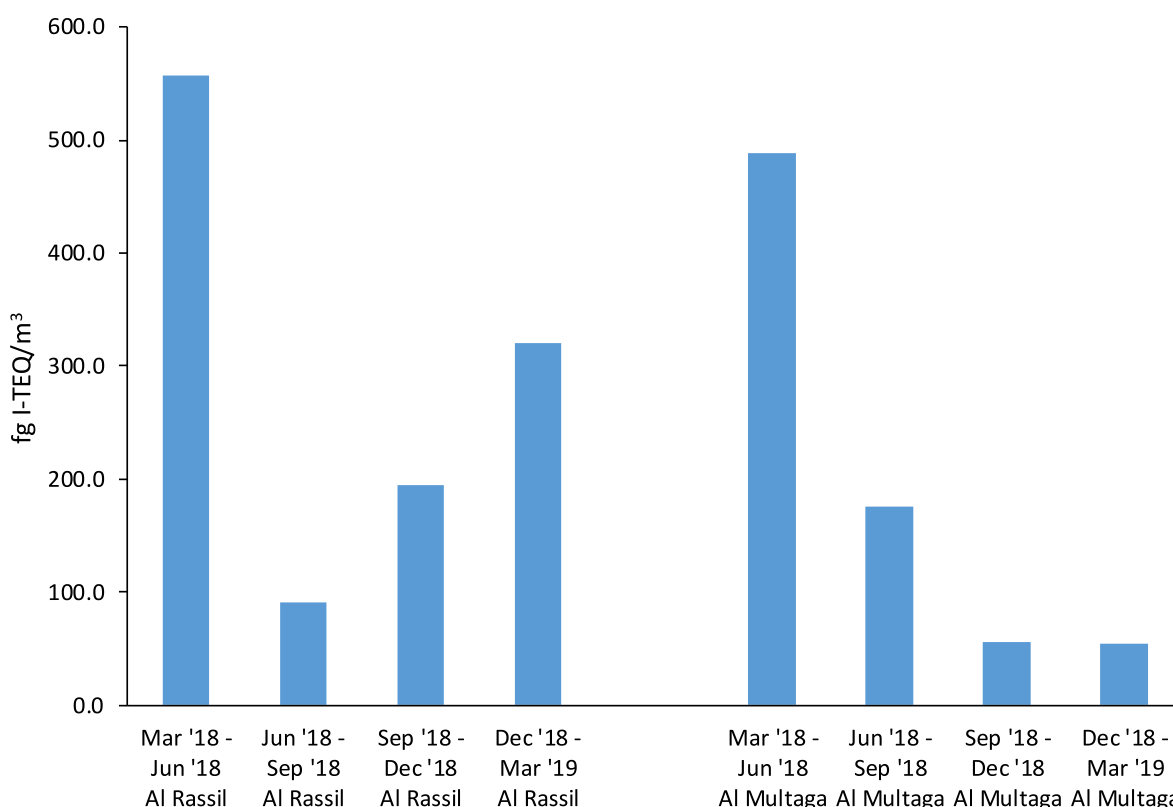


Fig. 5. Seasonal variations in concentrations of polychlorinated dibenzo-p-dioxins and dibenzofurans at two locations in Oman between March 2018 and March 2019.

m⁻³) but lower than values observed at Baraki (590–780 fg I-TEQ m⁻³) in northern Algeria (Moussaoui et al., 2012), Cuba (2042 fg WHO₉₈ TEQ m⁻³) and some sites in Africa (Bogdal et al., 2013). The only other PCDD/F study in Kuwait (Martinez-Guijarro et al., 2017), carried out using high volume air samplers at a site close to the remote sampling location in this study, reported TEQ values ranging between 34 and 586 fg I-TEQ m⁻³ (median, 95 fg I-TEQ m⁻³) which are similar to the values in this study. Higher concentrations than those observed in Kuwait and Oman have been reported previously at an urban site in Catalonia, Spain (1196 fg I-TEQ m⁻³ (Abad et al., 2007)), a background site in Catalonia, Spain (674 fg I-TEQ m⁻³ (Parera et al., 2018)), an industrial site in the

Malopolska region, Poland (2900 fg I-TEQ m⁻³ (Umlauf et al., 2010)), and in Guangzhou, China (3475 fg I-TEQ m⁻³ (Deng, 2013)) using high volume air samplers.

TEQ of \sum_{17} PCDD/F range from 52 fg I-TEQ/m³ to 229 fg I-TEQ/m³ with an average value of 123 fg I-TEQ/m³ in Istanbul (Gunes et al., 2014). A study in Istanbul-Turkey reported PCDD/F concentrations ranging from 0.5 ± 0.2 to 27.9 ± 8.4 fg I-TEQ m⁻³ in high-vol sampled ambient air samples (Ercan and Dinçer, 2016). In another study conducted in Istanbul, average gas phase concentrations of PCDD/F compounds at Davutpasa, Yildiz, and Fenertepe locations reported to be 175 fg m⁻³, 91 fg m⁻³, and 133 fg m⁻³ in samples collected between 2011

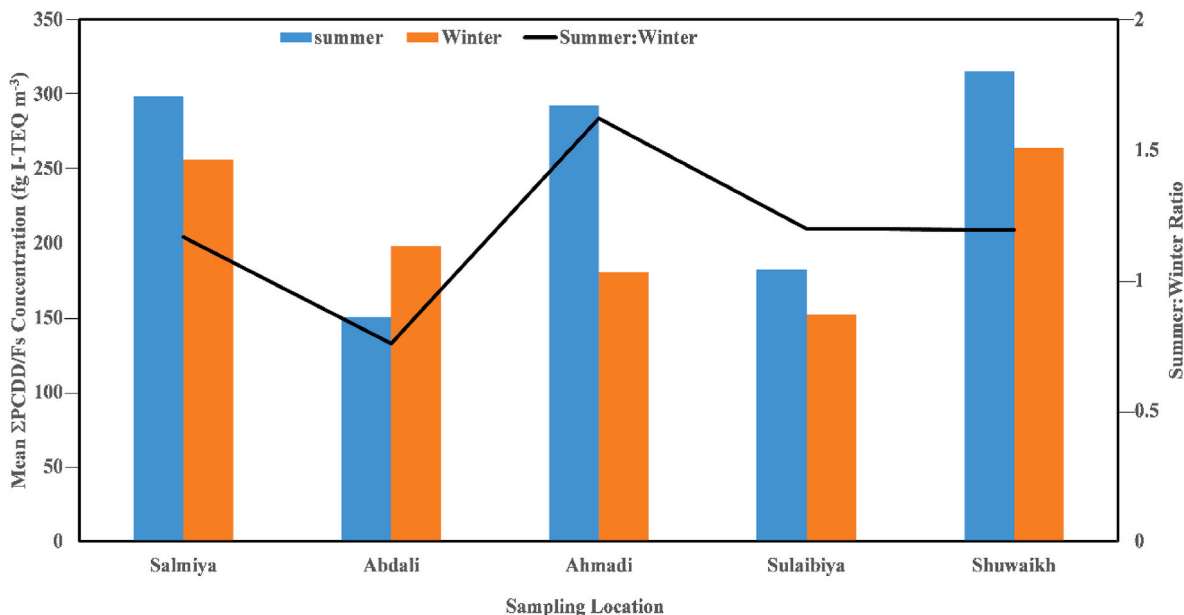


Fig. 6. Mean Averaged Summer and Winter ΣPCDD/F Concentrations and the Summer:Winter Ratio at five Locations across Kuwait between January 2018 and December 2018.

and 2013 (Sarial et al., 2015) whereas average concentrations of PCDD/F compounds were found to be (156 fg I-TEQ/m³), (65 fg I-TEQ/m³), and (40 fg I-TEQ/m³) for the same locations in samples collected in 2011–2012 (Gunes and Saral, 2014). In the current study, ΣPCDD/F concentration ranged between 19 and 75 fg I-TEQ/m³ in BUTAL-Bursa and 20–30 fg I-TEQ/m³ in Uludag-Bursa which were similar or lower than those reported in previous studies conducted in Turkey.

3.3. Homolog profiles

The homolog profiles for all sampling sites in the various countries are given in Fig. 7. The profiles at each site across the Middle East suggest that combustion/thermal sources are the predominant contributors to the PCDD/F in the atmosphere. Since there are marked differences at some of these sites, which appear to indicate that the

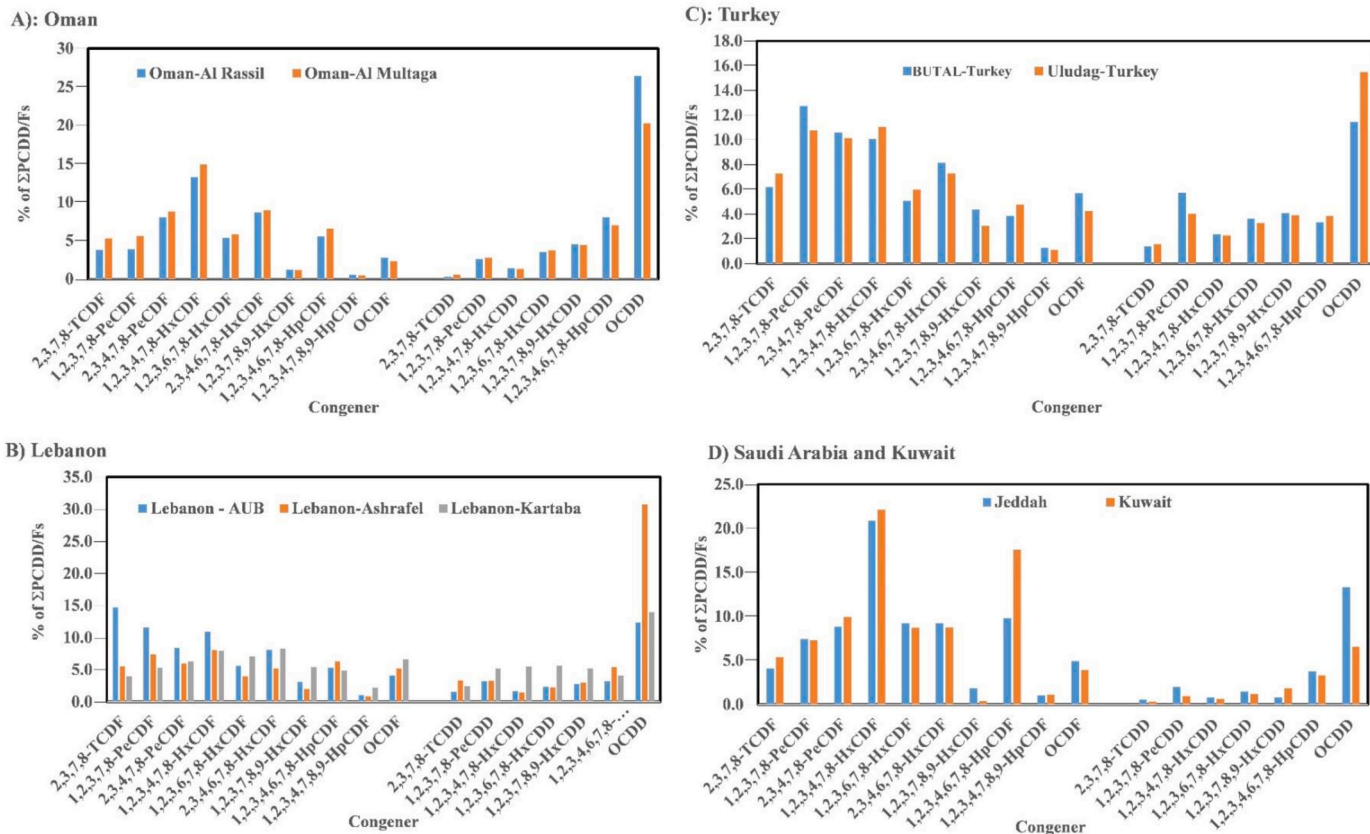


Fig. 7. Profiles of 2,3,7,8-substituted PCDD/F in air samples from five Middle Eastern countries between January 2018 and December 2018.

combustion sources contributing to the profiles are unique, the profiles are discussed on a country/site-specific basis.

The homolog profiles in samples collected from sites in Kuwait were dominated by PCDFs, comprising 85 and 77%, respectively. In Kuwait, the profile was dominated by 1,2,3,4,7,8-HxCDF contributing ca 22%, followed in decreasing order by 1,2,3,4,6,7,8-HpCDF (17%), 1,2,3,4,6,7,8-HpCDF \approx 2,3,4,6,7,8-HxCDF \approx 1,2,3,7,8,9-HxCDF (ca 9%) and OCDD \approx 2,3,4,7,8-PeCDF (7%). In Jeddah, the congener profile was also dominated by 1,2,3,4,7,8-HxCDF contributing ca 21%, followed in decreasing order by OCDD (13%) and 1,2,3,4,6,7,8-HpCDF, 2,3,4,6,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, and 2,3,4,7,8-PeCDF all contributing ca 9% each to the total PCDD/F. When the profiles at the sampling locations in these two countries are compared with the fingerprints reported by Cleverly et al. (1997), it can be concluded that the profile is indicative of a combination of incineration sources like municipal solid waste incineration, open burning, and unleaded fuel combustion as the main contributors to the profile.

The congener profiles at Ashrafiieh and Qartaba in Lebanon are broadly identical but different from those of samples collected at the American University of Beirut (AUB) Campus. At the Ashrafiieh and Qartaba locations, the profile was dominated by the OCDD, contributing 31% and 14%, respectively, followed by the HxCDF and PeCDF homologs. The profiles at these sampling locations indicate of the combustion of industrial wood and diesel emissions from vehicles (Cleverly et al., 1997). The dominant congener in samples collected from the campus of the American University of Beirut was 2,3,7,8-TCDF (15%) followed in decreasing order by OCDD (12%), 1,2,3,7,8-PeCDF (12%), and 1,2,3,4,7,8-HxCDF (11%). This profile is unique, but identical to profiles reported from emissions from a cement kiln burning non-hazardous waste by Cleverly et al. (1997). The University is located close to a hospital incinerator and is in an area with heavy vehicular traffic and impacted by open waste burning considered the major PCDD/PCDF source in Lebanon (2017). This profile may reflect the combination of these combustion sources rather than one specific source.

The homolog profiles in samples collected from sites in Turkey are similar to those in samples from Kuwait, with minor differences. The furans comprise 66% at both sites, with the PeCDF and HxCDF dominating the mixture. The following congeners 2,3,4,7,8-PeCDF, 1,2,3,7,8-PeCDF, 1,2,3,4,7,8-HxCDF, and OCDF made identical contributions of ca 11% of the Σ PCDD/F at both sites. This congener pattern suggests a combination of potential sources, including open burning of wastes and unleaded fuel combustion (the Butal site is located along a major highway). Lube oil is commonly used as fuel in vehicles, municipal solid waste incineration, and/or secondary lead smelting may be supplying the air around the sampling locations (Shen et al., 2005).

It is widely reported that characteristic congener and homolog profiles of PCDD/F are associated with different sources (Cleverly et al., 1997). Also, degradation processes in air take place to some extent, such as degradation by UV and hydroxyl radicals where lower chlorinated PCDD/PCDFs are degraded faster compared to higher chlorinated congeners, in particular OCDD (Choi et al., 2000). It is worth noting that in many of the samples, the 1,2,3,4,7,8-HxCDF had two to three times higher concentrations compared to 1,2,3,6,7,8-HxCDF. This is not observed in combustion or other thermal sources like the metal industry, where these two congeners have similar concentrations. The levels of 1,2,3,4,7,8-HxCDF and 1,2,3,6,7,8-HxCDF are generally identical or only somewhat higher (less than a factor of 2) for 1,2,3,4,7,8-HxCDF compared to 1,2,3,6,7,8-HxCDF in similar studies across the globe (Klima et al., 2020; Lohmann et al., 1999). High levels of 1,2,3,4,7,8-HxCDF is characteristic of processes which involve elemental chlorine, such as the chloralkaline process or the production of bleaching powder in Leblanc processes with a 1,2,3,4,7,8-HxCDF to 1,2,3,6,7,8-HxCDF ratio of 5 or 6 (Balzer et al., 2008; Rappe, 1994; Xu et al., 2000). This indicates that in the region, some impact of a source with elevated 1,2,3,4,7,8-HxCDF exists. The higher 1,2,3,4,7,8-HxCDF to 1,2,3,6,7,8-HxCDF ratio was also detected near the oil production/refinery

area in Kuwait (up to a factor of 2.6), which was likely impacted by this industrial facility (Martinez-Guijarro et al., 2017).

4. Conclusion

Atmospheric levels of PCDD/F were investigated at 12 sites across four Middle Eastern countries. Validated polyurethane foam passive air samplers were utilised for concurrent monitoring of PCDD/F in Kuwait, Turkey, Lebanon and Oman in order to establish baseline concentration levels across the region. The highest concentrations of Σ PCDD/F were detected in Oman (Al-Rassil Industrial area: 557 fg I-TEQ m^{-3} ; Al-Multaga: 489 fg I-TEQ m^{-3}). Sampling in Kuwait also showed elevated concentrations levels ranging from \sim 150 fg I-TEQ m^{-3} (Shuwaikh) to \sim 250 fg I-TEQ m^{-3} (Salmiya and Abdali). Among the countries where samples were obtained, Kuwait is the only member of the GAPS network, and this study is a first step toward filling the void of PCDD/F data shortage in the Middle East. Based on the results of the previous studies, a strong emphasis should be placed on improving the comparability of data collected by different POP monitoring networks to connect data gaps between regions. Therefore, the current regional monitoring study can act as a key research activity to pioneer routine monitoring activities in the region. Hence, similar monitoring activity to be conducted every 3 or 4 years for selected POPs appears to be a particular need for the region.

Credit author statement

Bondi Gevao: Contributed to the conception and design of the study, participant recruitment and performed data analysis and interpretation, Karell Martinez-Guijarro: Contributed to sample preparation, sample analysis, data analysis and interpretation, technical writing, Perihan Binnur Kurt-Karakus: Contributed to data analysis and interpretation, technical writing, Roland Weber: Contributed to interpretation and technical writing, Askin Birgul: Conducted sample collection in Turkey and technical writing, Carol Sukhn, Divya Krishnan, Smitha Rajagopalan, Hassan Alshemmari, Mariam Hajeyah, Majed Bahloul, Mohamed I. Orif: Contributed to sample collection, data analysis and technical writing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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