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Airborne Dioxins, Furans and Polycyclic Aromatic Hydrocarbons Exposure to Military Personnel in Iraq

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Abstract

Objectives—The objective was to use ambient polycyclic aromatic hydrocarbon (PAH), polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) concentrations measured at Joint Base Balad in Iraq in 2007 to identify the sources of these species and their spatial patterns.

Methods—The ratios of the measured species were compared to literature data for likely emission sources. Using the multiple site measurements on specific days, contour maps have been drawn using inverse distance weighting (IDW).

Results—These analyses suggest multiple sources including the burn pit (primarily a source of PCDD/PCDFs), the transportation field (primarily as source of PAHs) and other sources of PAHs that include aircraft, space heating, and diesel power generation.

Conclusions—The nature and locations of the sources were identified. PCDD/PCDFs were emitted by the burn pit. Multiple PAH sources exist across the base.

Keywords

burn pit; polychlorinated benzo-p-dioxins; polychlorinated dibenzofurans; particulate matter; polycyclic aromatic hydrocarbons

During the last decades, a large number of epidemiological studies have provided strong evidence of a direct association between exposure to ambient air pollutants and adverse effects on public health, including carcinogenic effects,[1,2,3] respiratory,[4,5,6] and cardiovascular diseases.[7,8,9] In 2013, outdoor air pollution and airborne particulate matter were classified by IARC as human carcinogens (Group 1).[10]

Open burning (OB) results in substantial emissions with emission factors (mass pollutant per mass fuel) much larger than well-controlled combustion sources,[11] such as many mobile and industrial sources. OB emissions can be highly variable. Their emission factors and chemical compositions are largely unpredictable because OB may involve a variety of sources and

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There are no conflicts of interest

fuels, e.g. various refuse waste and household trash burning,[12] accidental, yard and construction debris fires,[11] shredded tires,[13] landfill fires,[14] and e-waste recycling.[15] Estrellan and Iino[16] have reviewed the emissions of toxic species such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) and report that open burning of domestic and industrial wastes leads to the formation of toxic pollutants especially in the presence of polymeric materials. Emissions from open burning of plastics, tires, and electronic wastes, along with household scraps such as paper products and yard trimmings produce localized exposures to these pollutants.

Beside the large number of potential civil sources, military OB (MOB) emissions occur during deployment to war zones.[17] MOBs are the unavoidable consequence of: (i) large volumes of wastes generated from military exercises, operations, base management, personnel provision, vehicle, weapon and ammunition use; (ii) the unavailability of other suitable waste-disposal options and related infrastructures and facilities at forward operating bases, and (iii) the need for cost-effective elimination of hazardous wastes. The most common practice to routinely eliminate primary solid-wastes on US bases during Iraqi and Afghanistan conflicts was uncontrolled burning of the wastes in open-air burn-pits (BPs). In BPs, large amounts of refuse materials of different composition and hazardous potential were set in piles, rows, or holes in the ground and ignited by using raw fuels (e.g., jet fuel) as ignition accelerant. The combustion in BPs were mostly uncontrolled neither providing optimal burning conditions nor controlling the excess air ratio. Under such circumstances, smoldering combustion may occur and become dominant. As a consequence, large amounts of soot and unburned byproducts were released in the atmosphere along with aerosolized materials. As a consequence of BP practices, many military personnel and people living in the vicinity of the bases were exposed to smoke along with the other emissions from various sources (motor vehicles, aircraft, etc.). The use of BP was restricted in 2009 and their use in Iraq was gradually phased out at the end of 2010. However, they were also used in Afghanistan, where 197 burn pits were operating as of January 2011.[17]

Veterans returning from the conflicts in Iraq have reported health issues[18,19,20] that may be the consequence of exposures to the high airborne dust levels in arid environments, toxic emissions, extreme physical demands and/or combat situations (explosive blast, etc).[21] In 2008, the U.S. Department of Defense (DoD) claimed that there was no evidence that exposure to BP emissions created a long-term health risk for deployed personnel and stated that it posed an “acceptable health risk” based on the contaminant levels measured.[22] A number of studies have subsequently investigated the potential health consequences of exposure to BP smoke.[23,24,25,26,27,28] Falvo et al.[29] summarized the existing published data related to inhalational exposures and the respiratory health of service members deployed to Iraq. A substantial fraction of deployed personnel reported respiratory symptoms, illnesses and conditions. Krefft et al.[30] report that deployment to southwest Asia is associated with a wide range of respiratory disorders although they do not specifically relate any of the observed conditions to specific exposure sources. However, Abraham et al. reported that medical encounter rates are not uniquely associated with the BP practices.[31] Smith et al.[32] found no increased risks for new-onset respiratory symptoms if deployed within 3 miles of burn pit. Conlin et al.[33] found that burn pit exposures were not consistently associated with an increase in birth defects or preterm birth in infants of active-duty military personnel. Despite studies reporting that the BP practice do not constitute an elevated risk for human health, this conclusion is still heavily debated, mostly because toxicological, epidemiological and clinical data are not yet adequate to reliably evaluate the prevalence or severity of adverse effects of inhalational exposures to BP exhausts in military personnel.[34,35]

This study analyzes air pollution data collected at the Joint Base Balad (JBB), which hosted the largest operating burn-pit on U.S. bases during the Iraq War. Daily samples of particulate- and gaseous-phase organics were collected between January and November 2007 at five sites within the base perimeter or downwind of the burn-pit. The total (particulate + gaseous phase) concentrations of polychlorinated dibenzo-dioxins (PCDDs), -furans (PCDFs) and polycyclic aromatic hydrocarbons (PAHs) were quantified. Data were collected by DoD under environmental monitoring and health studies. This Enhanced Particulate Matter Surveillance Program was reviewed by the National Research Council (NRC).[36] The resulting chemical data were processed along with micro-meteorological parameters to assess the effect of BP burning upon air quality and to identify the potential sources of dioxins and PAHs in the ambient air at JBB.

METHODS

Study area and burn-pit features

JBB (33.95 N; 44.35 E; 50 m. a.s.l.), formerly called Logistics Support Activity (LSA) Anaconda, was the second largest US Base in Iraq (Figure 1). It spread over 25 km^2 and lies in a relatively flat region approx. 65 km North to Baghdad (~7.2 million inhabitants in 2011), about 19 km WNW to Balad (~250,000 inhabitants) and 1.5 km from the Tigris River. The area surrounding the base is mainly used for agriculture. The airfield (ICAO code: ORBD) was served by two ~3.5 km-long runways and hosted many military, housing, medical, service and other transport-related installations.

During the Iraq War, JBB hosted ~25,000 military, civilian, contractors and coalition personnel. DoD has estimated that about 3.6 to 4.5 kg day^{-1} of waste was typically produced by a soldier deployed at forward operating bases.[17] Using this assumption, about 90 to 180 tons of waste were produced per day at JBB. Beginning in 2003, a large BP was used to eliminate the solid waste produced by the base and related military operations. The BP was ~4 ha-wide and was located at the NE corner of the base, approx. 1.5 km from the closest housing area and approximately 500 m from a transportation field mainly used as large vehicle inspection station (Figure 1). It was estimated that in the early stages of troop deployment ~2 tons/day of waste was burned daily, but this amount likely reached up to 200 tons per day in 2007.[37]

Two incinerators were operational from autumn 2007 reducing the volume burned at BP by a factor of 2.[22] However, it is not clear if these “incinerators” were two-stage incinerators or air curtain burners.[38] JBB waste was mainly composed of a heterogeneous mixture of food waste, plastic (e.g., bottles, Styrofoam trays, meals-ready-to-eat packages, rubber), human waste, shipping and packaging materials, wood (e.g., treated and untreated pallets), chemicals (paints, solvents), oil, grease, petroleum, and metal/aluminum items (e.g., cans, wires), and was ignited with jet fuel. However, it was reported that electronics, tires, batteries, medical waste and expired pharmaceuticals were also occasionally burned.[16,**Error! Bookmark not defined.**] The typical waste composition at U.S. bases was simulated and analyzed by Woodall et al.[39] Aurell et al.[40] reported that emission factors at simulated BP are in general higher during smoldering conditions than during flaming for many chemicals. DoD characterized the waste burned at BP in Iraq as "municipal waste" with a raw composition of 5–6% plastics, 6–7% wood, 3–4% miscellaneous non-combustibles, 1–2% metals, and 81–84% combustible materials.[22]

Experimental

Samples were collected at five sampling sites (Figure 1): (i) mortar pit (MP), selected as a "background" site because of frequently upwind of BP emissions; (ii) guard tower (GT), a location outside the base perimeter and downwind of BP during prevailing wind regimes; (iii) transportation field (TF), located at a site close to BP but also potentially affected by emissions from diesel-powered heavy trucks. Between the BP and the TF site, there is a landing area for helicopters. The last two sites were placed in "living areas" of JBB: (iv) H-6 housing (H6) and (v) contingency aeromedical staging facility (CASF, CA). All living quarters and most work places were equipped with a heating and air conditioning unit. Electricity was generated by a number of diesel engine generators distributed around the base that operated continuously.

Samples were collected and analyzed according to US-EPA TO-9A and TO-13A methods. Gaseous compounds and PM_{10} (particles with aerodynamic diameter of 10 μm or less) were collected using high volume PS-1 air samplers (Tisch Environmental) equipped with 102 mm Ø quartz fiber pre-filters as media followed by XAD-2 polymeric adsorbent foam plugs. Samples were stored in clean Petri slides until analysis.

The semi-volatile nature of analyzed organic compounds drives their partitioning between the gases- and particulate-phases. The changes in relative partitioning of many congeners caused by atmospheric temperature may have major impacts on the outcomes of analytical results and may give artificial associations of congeners that are related more to their vapor pressures than to their sources. For this reason, the total (gaseous + particulate phases) concentrations are presented in this study: the filters and adsorbent cartridges were combined, solvent-extracted, concentrated, cleaned up using column chromatography to remove potential interferences and analyzed by HRGC-HRMS in the selected ion mode. Target compounds included 17 PAHs, naphthalene (NAP), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benz[*a*]anthracene (BaA), chrysene (CHR), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), benzo[*e*]pyrene (BeP), indeno[1,2,3-*cd*]pyrene (IP), dibenz[*a,h*]anthracene (DBahA), benzo[*g,h,i*]perylene (BghiP), 7 dioxins, 1,2,3,4,6,7,8-HeptaCDD, 1,2,3,4,7,8-HexaCDD, 1,2,3,6,7,8-HexaCDD, 1,2,3,7,8,9-HexaCDD, 1,2,3,7,8-PentaCDD, 2,3,7,8-TetraCDD, OctaCDD, and 10 furans, 1,2,3,4,6,7,8-HeptaCDF, 1,2,3,4,7,8,9-HeptaCDF, 1,2,3,4,7,8-HexaCDF, 1,2,3,6,7,8-HexaCDF, 1,2,3,7,8,9-HexaCDF, 1,2,3,7,8-PentaCDF, 2,3,4,6,7,8-HexaCDF, 2,3,4,7,8-

PentaCDF, 2,3,7,8-TetraCDF, OctaCDF. Suitably labeled, internal standards were used to quantify the compounds and assess recoveries. Data below the detection limits were set to DL/2.

Some micro-meteorological data were simultaneously recorded. Data recorded at the station were considered as representative of the weather parameters over the whole base, and include air temperature, atmospheric pressure, and wind speed and direction.

RESULTS

The analytical data is summarized in Figure 2 as box-plots while Table 1 lists the average, minimum and maximum concentrations of individual congeners and their sums (ΣPCDDs, ΣPCDFs and ΣPAHs for dioxins, furans and PAHs, respectively). Data in Table 1 are presented for the overall study period (2007) as well as split in winter-spring (January, February, April) and fall (October, November) to display the differences in concentrations. These differences may be due to: (i) the operation of two new incinerators in the fall 2007, which reduced by approximately 50% the volume of waste burned at BP and (ii) the effects of differing weather conditions. Figure 1 also presents the wind roses during the 4 seasons computed for the 2007 data. The prevailing wind direction is from the NNW. For this wind regime, GT is the most likely to be affected by the dispersion of BP plumes, while MP is generally upwind of the main JBB sources.

Generally, the concentrations of PCDD/Fs were found lower at MP and higher at GT and TF. The annual ΣPCDDs + ΣPCDFs average concentrations at the five sites followed the order (in pg m^{-3}): GT (18.6)> TF (12)> H6 (6.6)> CA (6.43)> MP (3). The average relative fraction of PCDD/Fs were almost identical at all sites (Figure 2) when considering the analytical uncertainty (±30%) and were dominated by OctaCDD (16%), 1,2,3,4,6,7,8-HeptaCDD (15%), 1,2,3,4,6,7,8-HeptaCDF (14%), 1,2,3,4,7,8-HexaCDF (13%), while other congeners accounted for less than 6% in mass concentration.

The lower annual average levels of ΣPAHs were recorded at MP. The highest concentrations of ΣPAHs were found at TF and followed the order (in μg m^{-3}): TF (0.64)> CA (0.43)> H6 (0.35)> GT (0.28)> MP (0.27). The ΣPAHs were overwhelmingly dominated by naphthalene, accounting for about 70-78% of all PAH mass at all sites (Figure 2), followed by other lighter congeners: PHE (7-10%), FLU (3-4%) and ACY (2-4%). The more carcinogenic congeners (BaP, DBahA) accounted for less than 1% in mass.

Differences in compound concentrations at the sampling sites may provide some insights about the potential sources, especially when also considering their closeness to the probable local sources and the variability due to changes in atmospheric circulation patterns. Using this approach, differences in ΣPCDDs, ΣPCDFs and ΣPAHs were investigated for each site and over the entire study period. However, due to their proximity, their relative location with respect to probable sources (BP, transportation field, runways and base camps) and the low number of samples collected individually, H6 and CA were combined into a single group.

Since Shapiro-Wilk tests applied to the data for each site revealed that only ΣPAHs values at GT were normally distributed at $p<0.05$, the nonparametric Kruskal-Wallis ANOVA by ranks was selected to test the significance of differences among sites. These results indicated that only the pairs MP-GT and MP-TF are significantly different at $p<0.05$ for ΣPCDD and ΣPCDF. Considering that prevailing winds over the study period blew from NW sectors, the results clearly indicate a probable strong source upwind of GT and TF, which is highly compatible with the burn-pit. For ΣPAHs, only the MP-TF pair showed significant different concentrations.

The PAH results indicated that it is distributed differently over the study area with respect to PCDDs/PCDFs. TF is located close to a station assigned to large vehicle inspections. Thus, it is potentially affected by emissions from heavy-duty diesel vehicles that are often idling for extended periods. It is also near the helipad. Diesel vehicles are known to be sources of airborne PAHs.[41,42,43,44,45] In addition to the ground vehicles, there were aircraft (fixed wing and helicopters) that are sources of PAH emissions.[46,47] Diesel generators were used at multiple sites across JBB to provide electricity. All of these systems (vehicles, aircraft, heating, and generators) use a single fuel, jet fuel JP-8. JP-8 is similar to the civilian jet fuel Jet-A, but had additives in it. The additives are a corrosion inhibitor/lubricity enhancer, a fuel system icing inhibitor, and a static dissipater additive. The measured concentrations indicate that multiple combustion sources were likely to have a role on the PAHs concentrations at JBB.

After October 2007, two incinerators were operative at JBB. As a consequence, the total amount of waste burned at BP was approximately halved.**Error! Bookmark not defined.** Table 1 shows the average concentrations between the pre- and post-deployment of the incinerators. The Kruskal-Wallis ANOVA was used to test the differences in PCDDs/PCDFs and PAHs concentrations between the two periods. Results do not show any significant ($p<0.05$) difference for ΣPCDDs and ΣPCDFs, while ΣPAHs were significantly higher at TF during November-December. As a matter of fact, this result drives two main considerations: (i) the deployment of the two incinerators did not result in a significant drop in emissions from BP or (ii) different weather factors linked to seasonality had an important role in the levels of pollutants and their dispersion into the atmosphere.

Toxic equivalencies

The toxic effects of the PAHs or the PCDDs/PCDFs are consistent with additive models (although affected by large variations) so that the relative toxic equivalency factors (TEFs) can be assigned for individual congeners and summed to produce total toxic equivalents (TEQs) relative to a reference compound. TEQs are computed by the sum of the products of the concentration of each compound multiplied by its TEF value. In this study, TEFs reviewed in 2005 by WHO[48] are used to calculate the TEQs for the PCDDs/PCDFs. The modified TEF lists provided by Nisbet and LaGoy,[49] Malcolm and Dobson,[50] and Larsen and Larsen[51] (1998) were used for PAHs.

Results are shown in Figure 2. The lowest TEQ values relative to 2,3,7,8-TCDD (ΣTCDD-TEQ) were found at MP and highest at GT and TF. The average ΣTCDD-TEQ concentrations at the five sites followed the order (in pg m^{-3}): GT (1.52)> TF (0.91)> CA (0.52)> H6 (0.51)> MP (0.25). The homologues which mostly contributed to ΣTCDD-TEQ were almost identical at all the sites (Figure 2) and were dominated by 1,2,3,7,8-PCDD and

2,3,4,7,8-PCDF (21%), 1,2,3,4,7,8-HeCDF (16%), 2,3,7,8-TCDD (8%), while other homologues accounted for less than 7% in total TEQ. For PAHs, the highest TEQs relative to BaP (BaP-TEQs) were found at TF (4.2 ng m^{-3}), followed by CA (3.6) and GT (3) and were dominated by BaP (~50%). The high average BaP-TEQ recorded at CASF was unexpected. However, a possible explanation may be linked to the low number of collected samples (N=5) or the use of jet fuel (JP-8) for space heating and electricity generation.

Comparisons with known profiles

Ratios among the measured compounds can be useful in preliminarily investigation into the most probable emission sources, when compared with known source profiles. Using this approach, diagnostic ratios (DRs) between concentrations of individual PAHs congeners are frequently used (e.g., Ravindra et al.[41] and references therein). There are limitations and weakness in using DRs for source identification. [52,53,54,55,56] The problems are linked to the differing chemical-physical properties of congeners, which may drive the environmental fate of these compounds after emission into the atmosphere. In fact, ambient temperature affects the different partitioning of congeners between gaseous- and condensed-phases and atmospheric oxidants (radicals, ozone, nitrogen dioxide) may deplete the most reactive congeners.

To overcome these issues, in this study DRs are critically interpreted in view of 4 key points: (i) total (gaseous+ particle) concentrations were used to discharge the variability of ratios related to the gas–particle partitioning; (ii) samples were collected close to the most probable emission sources in the area (within ~3.5 km from BP, runways and transportation field) minimizing artifacts due to the chemical and photolytic depletion of the more reactive species; (iii) only the more conservative ratios were calculated, i.e., FLT/(FLT+PYR), ANT/(ANT + PHE), BaA/(BaA + CHR) and IP/(IP + BghiP) according to the literature; and (iv) ratios recorded at JBB were matched to profiles analyzed with comparable equipment and lab methods to avoid artifacts related to analytical uncertainties. In particular, PCDDs+Fs and PAHs profiles were derived from: simulated deployed US military waste emissions;[39] legacy (1985 6.2L and 1993 10L) heavy-duty diesel engines at transient on-road settings;[43] common in-use heavy-duty diesel engines (1998 11L) without after-treatment devices at cruise, urban, and idling speeds;[44] modern diesel engines (2008 6.7L and 2008 6.4L) with and without exhaust gas after-treatment at average conditions;[42,45] various aircraft turbofan engines.[47,57]

Figure 3 shows the ratios calculated over the JBB samples and the profiles reported in the literature: the ratios related to the simulated deployed military waste emissions are shown as average and relative standard errors (dashed lines). The FLT/(FLT + PYR) vs. ANT/(ANT + PHE) and IP/(IP + BghiP) vs. BaA/(BaA + CHR) plots clearly show that profiles from the simulated military waste relate well for most of the samples collected at JBB. However, considering the uncertainties associated with the analytical methods, some DRs also match the profiles reported in the literature for diesel[39,42] and turbofan engines.[47,57]

The scatterplot of ΣPAHs vs. ΣPCDDs concentrations at JBB (Figure 3) revealed a null coefficient of determination ($R^2_{adj}\approx 0$). However, data are distributed along two main populations, showing that there were at least two main possible sources: one mostly linked to strong increases of PAHs and the other towards higher dioxins to PAHs ratios. The ratio between PAHs and PCDDs was thus also investigated. Figure 3 reports the scatterplot of the fraction of high molecular weight PAHs (5- and 6-rings) to ΣPAHs versus the fraction of dioxins on the PCDDs + PCDFs and reveals that data collected at JBB may fit with both simulated military waste burning and diesel emissions.

Although the data collected at JBB and profiles for simulated waste burning exhibit a good fit for the PCDDs to PCDDs + PCDFs ratio, Figure 3 also shows that there is not good agreements when considering the fraction of single PCDD/Fs homologues, i.e. 2,3,7,8-TCDD at JBB is well below the data reported in the literature. Supplemental Figure S1 shows the fraction of single homologues on PCDDs + PCDFs and reveals that data collected at JBB highly differ from data reported by Woodall et al.[39] for open burning of simulated military waste. In particular, fractions of 2,3,7,8-TCDD, 1,2,3,7,8-PentaCDD, 2,3,4,7,8-PentaCDF and 2,3,4,6,7,8-HexaCDF were significantly lower at JBB, while 1,2,3,7,8,9-HexaCDD, 1,2,3,4,6,7,8-HeptaCDD, OctaCDD, 1,2,3,4,6,7,8-HeptaCDF and 1,2,3,4,7,8,9-HeptaCDF were higher. In summary, considering the high variability of the data due to large analytical uncertainties associated with the dioxin analysis as well as the large variation in materials burned at BPs, the data from JBB generally demonstrate high emission for PCDD and PCDF enriched in chlorine. The difference can be explained by a number of factors: (i) the burning of materials with different composition at the BP; (ii) differing burning conditions; and (iii) the presence of other sources of PCDD/Fs at JBB. This latter point could be linked to PCDD/Fs emitted from mobile sources, such as heavy duty diesel vehicles at the station assigned to large vehicle inspections near TF. On the other hand, the potential role of emissions from military aircrafts and helicopters to PCDD/Fs was not evident: aircrafts are not recognized as sources of dioxins,[58] although the presence of chlorine in jet fuels or in ambient air may have a role in increasing the emissions from turbofan engines.

Spatial analysis

A preliminary qualitative investigation of the potential effects of BP was conducted by analyzing the spatial distribution of pollutants along with wind roses for all of the sampling days on which more than one site was sampled and analyzed. Each site was taken as representative of an area (~1 mile radius for buffer). The “mortar pit” (MP) site was selected as “background” because it was upwind of the other sites for the prevailing wind regime on almost all of the days on which samples were collected.

Since ΣPCDDs and ΣPAHs were simultaneously available at only three sites on a single monitoring day, a simple deterministic method was selected for the spatial interpolation (inverse distance weighting, IDW). Using IDW, the influence of neighboring points is diminished as a function of increasing distance d (d^2 in this case). All of the resulting maps are provided in Supplementary Figures S2-22.

Figure 4 presents the three maps showing the most commonly observed spatial distributions for the PAHs, PCDDs, and PCDFs. Minimal concentrations are typically observed at the northwest end of the base. This area is generally upwind of the BP and the TF emission areas. The PCDD and PCDF concentration patterns show high concentrations in the vicinity of the BP. The PAH pattern is quite different being focused in the area between the TF and the housing areas (CASF and H-6 Housing). Given the northwesterly wind direction, the emissions from the TF and possibly the helipad plus the emissions from diesel generators and building heating are likely to be the major PAH sources with some contribution from the BP.

The distributions on February 8 (Figure S 5) show the higher PCDD and PCDF further to the SW, but it should be noted that the maximum concentration on this date is approximately 30x lower than on January 3. The PAH concentrations are more focused near the TF and again have a maximum concentration lower than January 3 although in this case by only a factor of 3.4. These findings suggest differences in the sources of the PCDD/PCDF and the PAH compounds. Comparing the PAH maps with those for the PCDD/PCDFs suggests that for most days, the BP is not a major source of PAHs relative to other sources on the base.

On April 11 (Figure S 9), the concentrations are highest at the northwestern end of the modeled domain. There are low wind speeds on this day with some southeasterly flow. However, this pattern is anomalous relative to the BP area that is expected to be the major source of the PCDD and PCDF species.

Examining the PCDD and PCDF distributions over all of the sampling days, it can be seen that their spatial distributions are virtually identical except for April 9 (Figure S 7) and 14 (Figure S 11). April 9 had southeasterly and southern winds and moderate concentration values. On April 14, there were also southerly winds. For the April 9 map, the PCDD pattern is consistent with BP emissions pushed northward by the southerly winds. However, the PCDF maps also show higher concentrations within the SE portion of the modeled domain. For April 14, the PCDF pattern is more consistent with BP emissions being transported northward while the PCDD concentrations are highest in the southeastern portion of the domain.

In conclusion, spatial analysis revealed that the origin of PCDDs/PCDFs is attributed to the burning of waste in the BP while the PAH pollution appears to be related to a variety of sources within JBB including motor vehicles, aircraft, electricity generation, and space heating. These maps could be used to provide an exposure assessment if time-activity data were available for personnel working in this area of the base. Certainly personnel working at the guard tower are much more likely to have been exposed to burn pit emissions than those personnel working elsewhere on the base. There appears to be much more general exposure to PAHs given their higher concentrations in the housing areas of the base within the modeling domain.

CONCLUSIONS

This study has investigated the distributions of PCDDs, PCDFs and PAHs at JBB during 2007 in view of the likely sources and their chemical and spatial characteristics. The main results can be summarized as follows:

- Very high levels of gas- + particulate-phase organics are recorded at JBB. The highest concentrations of PCDD/Fs are recorded at GT, a site downwind to the burn pit during prevailing wind regimes and, thus, potentially affected by dispersion of plume from open burning of military waste. Highest levels of PAHs are found at TF, a site heavily affected by emissions from heavy-duty diesel vehicles;

- No significant differences are found between January-April and October-November, i.e. no significant decreases in levels of compounds are observed after two incinerators were deployed at JBB;
- Ratios among PAH congeners and PAH to PCDD ratio revealed good agreements with data reported in the literature for the open burning of simulated military waste. However, ratios were also compatible (within the analytical uncertainty of data) with emissions from diesel engines. Ratios among single PCDD and PCDF species to the total PCDD/Fs concentrations showed a larger fraction of the more chlorinated homologues was found at JBB. This result can be likely attributed to the burned materials and/or conditions; and
- A preliminary spatial analysis of results coupled with the analysis of wind directions showed that the distribution of the PCDDs/PCDFs over the base are often highly related to the dispersion of BP plumes. However, spatial distributions not related to BP plumes were also observed on some days, mostly for PAHs. This result can be attributed to the presence of multiple sources of PAHs at JBB.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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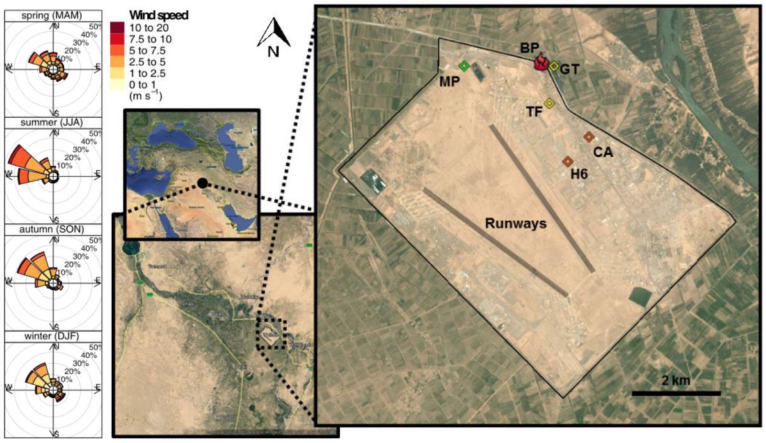


Figure 1.

Map of the study area and wind roses computed seasonally on 2007 data. JBB sites: burn-pit (BP); guard tower (GT); transportation field (TF); H6-Housing (H6); contingency aeromedical staging facility (CASF, CA).

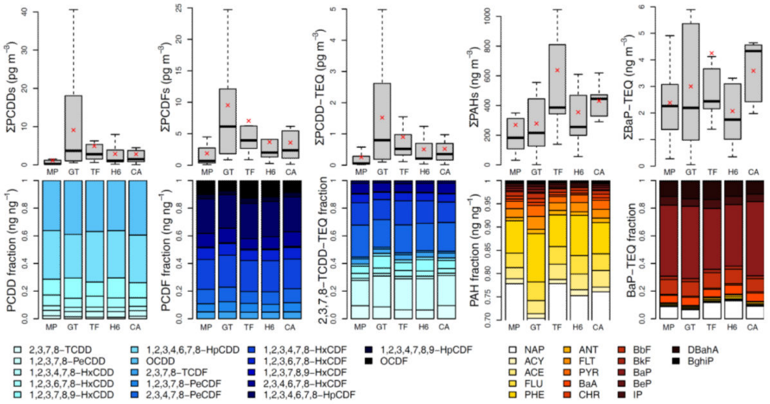


Figure 2.

Distributions of analytical results as box-plots (top) and average relative fractions of homologues and congeners (bottom). Box-plots: boxes represent inter-quartile ranges, black lines are medians, whiskers are quartiles ± (1.5*inter-quartile ranges), red crosses are arithmetical means. NAP is partially hidden on the PAH fraction plot because it represents about 70-78% of total mass concentration of PAHs. 2,3,7,8-TCDD-TEQ are computed according to TEQs proposed by Van den Berg et al.;[48] BaP-TEQ are computed on TEFs according to Nisbet and LaGoy[49] and Larsen and Larsen.[51]

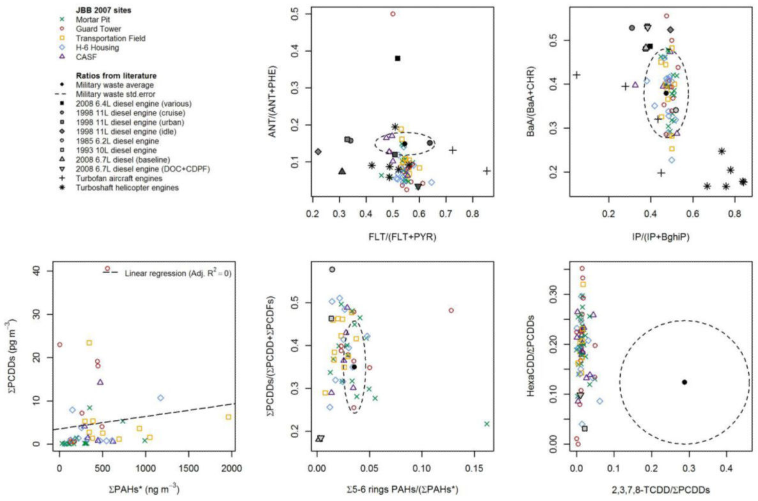
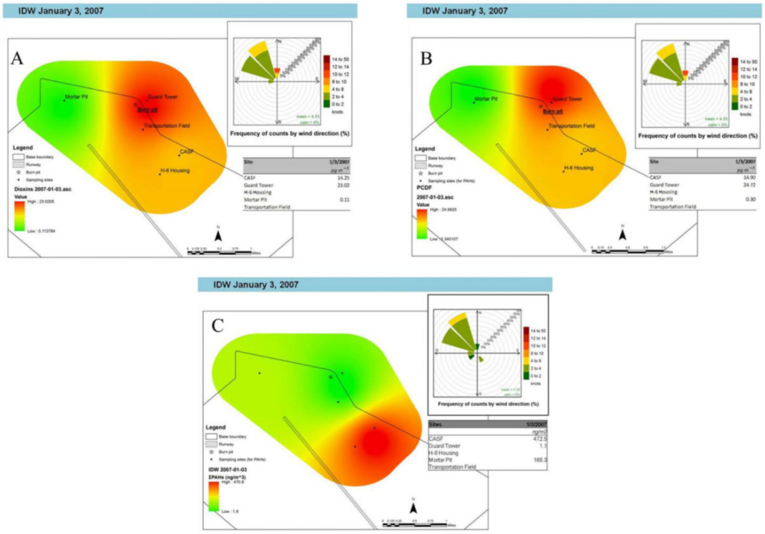


Figure 3.

Scatterplots between pairs of diagnostic ratios for PAHs (top); the regression between PCDDs and PAHs (bottom left) and mixed ratios among PCDDs, PCDFs and PAHs (bottom center and right). Data collected at JBB are colored. Data from literature are also reported for a comparison: simulated US military waste;[39] legacy heavy-duty diesel engines at transient on-road settings;[43] common in-use heavy-duty diesel engines without after-treatment devices at cruise, urban and idle thrusts;[44] modern diesel engines with and without exhaust gas after-treatment at average conditions;[42,45] various turbofan engines.[57]



Site	1/3/2007
	pg m⁻³
CASF	14.25
Guard Tower	23.02
H-6 Housing	
Mortar Pit	0.11
Transportation Field	

Site	1/3/2007
	pg m⁻³
CASF	14.90
Guard Tower	24.72
H-6 Housing	
Mortar Pit	0.30
Transportation Field	

Sites	1/3/2007
	ng/m3
CASF	472.5
Guard Tower	1.1
H-6 Housing	
Mortar Pit	165.3
Transportation Field	

Figure 4.

Interpolated maps for the sum of the PCDD compounds (A), PCDF compounds (B), and PAH compounds (C) on January 3, 2007.

Table 1

Average concentrations of dioxin/furan compounds at all the sites. Data are split to show the whole sampling campaign (2007) and the two periods without and with the use of the two incinerators (January, February, April: JFA; October-November: ON). Data below the detection limits (DLs) are estimated as DL/2.

PCDD/Fs (pg m^{-3})	TEF [a]	MP		GT		TF		H6		CA		All sites
		JFA	ON	JFA	ON	JFA	ON	JFA	ON	JFA	ON	2007
N		15	10	9	5	6	5	5	5	9	5	74
2,3,7,8-TetraCDD	1	0.03	0.01	0.09	0.21	0.07	0.04	0.05	0.01	0.04	0.06	0.06
1,2,3,7,8-PentaCDD	1	0.06	0.02	0.29	0.44	0.27	0.13	0.20	0.03	0.13	0.08	0.15
1,2,3,4,7,8-HexaCDD	0.1	0.05	0.02	0.23	0.38	0.22	0.08	0.16	0.02	0.11	0.06	0.12
1,2,3,6,7,8-HexaCDD	0.1	0.12	0.04	0.48	0.80	0.55	0.20	0.33	0.06	0.21	0.12	0.26
1,2,3,7,8,9-HexaCDD	0.1	0.19	0.05	1.24	1.47	0.71	0.27	0.75	0.08	0.37	0.19	0.48
1,2,3,4,6,7,8-HeptaCDD	0.01	0.55	0.20	2.07	4.33	2.45	1.03	1.66	0.31	1.10	0.71	1.27
OctaCDD	0.0003	0.46	0.35	1.79	6.68	2.43	1.09	1.68	0.40	1.15	1.04	1.43
2,3,7,8-TetraCDF	0.1	0.12	0.06	0.33	0.75	0.40	0.24	0.28	0.08	0.20	0.14	0.23
1,2,3,7,8-PentaCDF	0.03	0.14	0.06	0.48	1.07	0.51	0.38	0.31	0.11	0.24	0.22	0.31
2,3,4,7,8-PentaCDF	0.3	0.25	0.11	0.74	1.49	0.69	0.51	0.47	0.20	0.40	0.30	0.46
1,2,3,4,7,8-HexaCDF	0.1	0.52	0.23	1.78	2.95	1.93	1.14	1.26	0.39	0.90	0.54	1.05
1,2,3,6,7,8-HexaCDF	0.1	0.20	0.09	0.68	1.05	0.62	0.43	0.42	0.16	0.35	0.22	0.38
1,2,3,7,8,9-HexaCDF	0.1	0.01	0.01	0.03	0.10	0.04	0.06	0.03	0.02	0.03	0.03	0.03
2,3,4,6,7,8-HexaCDF	0.1	0.24	0.10	0.93	1.10	0.64	0.42	0.51	0.15	0.45	0.25	0.45
1,2,3,4,6,7,8-HeptaCDF	0.01	0.58	0.29	1.95	3.00	2.16	1.36	1.43	0.46	1.07	0.59	1.16
1,2,3,4,7,8,9-HeptaCDF	0.01	0.06	0.06	0.20	0.31	0.32	0.18	0.17	0.07	0.11	0.14	0.14
OctaCDF	0.0003	0.17	0.23	0.57	1.02	1.26	0.48	0.63	0.25	0.34	0.26	0.46
PCDD	—	1.47	0.70	6.18	14.31	6.70	2.83	4.82	0.92	3.11	2.24	3.76
PCDF	—	2.31	1.25	7.68	12.84	8.58	5.21	5.51	1.88	4.09	2.70	4.66
ΣPCDD-TEQ	—	0.33	0.14	1.22	2.06	1.12	0.64	0.81	0.21	0.59	0.40	0.68

[a] TEFs from Van den Berg et al.[48]

Table 2

Average concentrations of PAH compounds at all the sites. Data are split to show the whole sampling campaign (2007) and the two periods without and with the use of the two incinerators (January, February, April: JFA; October- November: ON). Data below the detection limits (DLs) are estimated as DL/2.

		MP		GT		TF		H6		CA		All sites
PAHs (ng m^{-3})	TEF [a]	JFA	ON	JFA	ON	JFA	ON	JFA	ON	JFA	ON	2007
N												
NAP	0.001	134	336	130	261	233	813	221	341	301	368	289
ACY	0.001	4.9	8.7	3.6	12.0	7.7	36.7	11.6	12.6	13.3	18.3	11.7
ACE	0.001	3.2	2.4	1.9	3.8	4.5	7.4	5.8	2.8	5.0	3.5	4.0
FLU	0.001	8.7	8.1	7.1	15.3	16.7	32.9	16.2	10.6	16.0	14.7	14.0
PHE	0.001	20	16	17	40	38	50	37	21	29	29	29
ANT	0.01	2.0	2.0	1.9	3.5	3.7	8.8	3.0	2.3	4.3	4.1	3.3
FLT	0.001	5.5	4.3	5.0	10.4	9.5	11.5	7.7	6.0	8.2	8.1	7.4
PYR	0.001	4.6	3.6	4.7	8.3	7.2	10.0	6.3	5.3	8.5	8.0	6.3
BaA	0.1	1.6	0.8	1.5	2.1	1.5	3.4	1.4	1.3	2.6	1.6	1.7
CHR	0.01	2.1	1.3	1.5	3.1	3.0	4.2	2.0	2.0	3.7	3.1	2.4
BbF	0.1	2.8	2.2	2.1	4.2	3.0	6.7	1.8	2.8	3.2	3.7	3.1
BkF	0.1	0.6	0.5	0.5	0.9	0.7	1.5	0.4	0.7	0.8	0.7	0.7
BaP	1	0.9	1.7	0.9	2.3	0.9	3.1	0.7	1.3	1.4	2.7	1.4
BeP	0.002[b]	1.5	1.2	1.2	2.4	1.6	3.6	1.0	1.4	1.8	1.8	1.7
IP	0.1	1.3	1.7	1.0	2.2	1.4	4.2	0.8	1.9	1.7	2.3	1.7
DBahA	1[c]	0.3	0.3	0.2	0.6	0.3	0.9	0.2	0.3	0.3	0.4	0.3
BghiP	0.01	1.4	1.7	1.1	2.2	1.5	4.4	0.9	2.3	1.9	3.8	1.9
ΣPAHs	—	196	392	182	374	334	1002	317	415	402	474	379
ΣBaP-TEQ	—	2.1	2.9	1.8	4.2	2.2	6.7	1.7	2.7	3.0	4.4	2.9

[a] TEFs for almost all the PAHs are adopted from Nisbet and LaGoy,[49] with the exception of:

[b] BeP from Larsen and Larsen[51]

[c] DBahA, the TEF of 1 was selected basing on Malcolm and Dobson.[50]