1 Bottom sediment as a source of organic contaminants in Lake Mead, Nevada, USA

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3

4 ABSTRACT

5 Treated wastewater effluent from Las Vegas, Nevada and surrounding communities' flow 6 through Las Vegas Wash (LVW) into the Lake Mead National Recreational Area at Las Vegas 7 Bay (LVB). Lake sediment is a likely sink for many hydrophobic synthetic organic compounds 8 (SOCs); however, partitioning between the sediment and the overlying water could result in the 9 sediment acting as a secondary contaminant source. Locating the chemical plumes may be 10 important to understanding possible chemical stressors to aquatic organisms. Passive sampling devices (SPMDs and POCIS) were suspended in LVB at depths of 3.0, 4.7, and 6.7 (lake bottom) 11 12 meters in June of 2008 to determine the vertical distribution of SOCs in the water column. A 13 custom sediment probe was used to also bury the samplers in the sediment at depths of 0-10, 10-14 20, and 20-30 cm. The greatest number of detections in samplers buried in the sediment was at the 0-10 cm depth. Concentrations of many hydrophobic SOCs were twice as high at the 15 16 sediment-water interface than in the mid and upper water column. Many SOCs related to wastewater effluents, including fragrances, insect repellants, sun block agents, and phosphate 17 flame retardants, were found at highest concentrations in the middle and upper water column. 18 19 There was evidence to suggest that the water infiltrated into the sediment had a different 20 chemical composition than the rest of the water column and could be a potential risk exposure to

21 bottom-dwelling aquatic organisms.

24 **1. Introduction**

25 In recent years, there has been an abundance of research determining the presence of synthetic 26 organic compounds (SOCs) in environmental waters, especially those receiving treated effluents 27 from municipal wastewater treatment plants (WWTPs). These studies generally focus on point 28 source impacts and the lateral distribution of SOCs downstream of known point sources. 29 Although the hydrodynamics within a flowing body of water (i.e., a stream or river) can be 30 complex, it is generally intuitive that SOCs will continue to move downstream until they are 31 removed by attenuation processes, such as sorption or degradation. 32 33 Less is known about the vertical distribution of SOCs in deeper large bodies of water (lakes) 34 which receive WWTP effluents. Dilution of WWTP effluent streams and the lack of sufficiently 35 sensitive analytical detection methods can limit the ability of researchers to determine the 36 presence of SOCs in the water column. Lake sediment is a likely sink for many SOCs; however, 37 partitioning between the sediment and the overlying water could result in the sediment acting as 38 a secondary contaminant source.

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40 The cities of Las Vegas, North Las Vegas, Henderson, and metropolitan Clark County, Nevada, 41 discharge treated wastewater effluent from three WWTPs into Las Vegas Wash (LVW) and 42 subsequently into the Lake Mead National Recreation Area at Las Vegas Bay (LVB, Fig. 1). 43 Lake Mead is the largest man-made reservoir by volume in the United States and was created in 44 1935 by the construction of Hoover Dam on the Lower Colorado River. Although the treated 45 effluent in LVW only accounts for a historical average of 1.4% of the total water flow into Lake 46 Mead, the presence of the SOCs in this effluent may be biologically important (Benotti et al.,

47 2010). Evidence of endocrine disruption in common carp (*Cyprinus carpio*) sampled from both
48 LVW and LVB provides a potential link between SOCs present in the WWTP effluents and
49 declining health of aquatic species in LVB (Bevans et al., 1996; Patiño et al., 2003).

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51 Rosen et al. (2010) studied the lateral and vertical gradients of SOCs throughout Lake Mead in 52 March of 2006 using passive sampling devices. Semipermeable membrane devices (SPMDs) 53 and polar organic chemical integrative samplers (POCIS) were deployed in LVB to determine 54 the vertical distribution of SOCs entering Lake Mead from LVW. Their study found that the 55 highest concentrations of hydrophobic SOCs were present nearest the lake bottom. This finding 56 along with data showing elevated water temperatures and higher total dissolved solids 57 concentrations along the lake bottom indicated that the effluent plume from LVW was moving 58 along the lower portion of the water column.

59

LaBounty and Burns (2005) determined that the water from LVW will move through LVB and
into Lake Mead at different vertical positions at different times of the year. During the winter
months, water from LVW moves across the bottom of the lake. As water temperatures increase,
the LVW plume goes through an unstable period in the spring and then moves across the lake's
surface in the summer.

65

The potential of lake sediments acting as a source of bioavailable contaminants has implications on the health of bottom-dwelling aquatic organisms. In order to distinguish between contaminants originating from the LVW plume and the contaminants associated with the sediment infiltrated water, two criteria had to be met: 1) the LVW plume needed to be near the

surface of the lake and 2) monitoring techniques which could distinguish between the available
contaminants in the infiltrated water and sediment-bound contaminants needed to be used.

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73 The use of passive samplers for measuring loosely bound organic chemicals in sediments and 74 sediment infiltrated water has been widely reported (De Jonge and Rothenberg, 2005; Namieśnik 75 et al., 2005; Huckins et al., 2006; Ouyang and Pawliszyn, 2006). SPMDs have been buried in 76 direct contact with sediments to measure the fraction of bioavailable SOCs (Rantalainen et al., 77 2000; Ya-xian et al., 2001; Williamson et al., 2002) and have been used in groundwater 78 monitoring studies deployed in wells (Vrana et al., 2005). In each of these cases, legacy 79 contaminants such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons 80 (PAHs) were targeted. This work is the first reporting of using SPMDs or POCIS to measure 81 emerging contaminants such as fragrances, pharmaceuticals, alternative flame retardants, 82 industrial and residential chemicals, in sediments and the first use of POCIS to measure SOCs in 83 sediment infiltrated water.

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All of the passive sampler/sediment studies mentioned previously had the limitation of needing direct access to the sediment in order to bury the passive samplers. Those approaches are not suited for lakes and other deep bodies of water where the sediment is inaccessible without specialized equipment. To overcome this limitation, a sediment probe was constructed which could be lowered from the surface and would insert the passive samplers into the sediment bed at defined depth intervals. The sediment probe described herein was capable of exposing SPMDs and POCIS to the sediment infiltrated water at three depth intervals.

93 **2. Experimental**

94 2.1 Chemicals of interest

95 A suite of 141 anthropogenic organic chemicals, not including the 140+ PCB congeners

- 96 measured to estimate the total PCB value, covering a range of hydrophobicities were targeted in
- 97 this work. Tables 1 and S1 list the chemicals sampled by the SPMD including legacy
- 98 organochlorine pesticides, PCBs, PAHs, and polybrominated diphenyl ether (PBDE) flame
- 99 retardants (Alvarez et al., 2008a; Petty et al., 2000). Tables 2 and S1 list the chemicals measured

100 in POCIS extracts including organic wastewater chemicals (OWCs, including fragrances, flame

101 retardants, plasticizers, personal care products, and industrial and consumer chemicals),

102 pharmaceuticals and illicit drugs (Alvarez et al., 2008b; Jones-Lepp et al., 2004). Extracts from

103 both SPMDs and POCIS were screened for estrogens and estrogen-mimicking chemicals using

104 the yeast estrogen screen (YES) (Alvarez et al., 2008a).

105

106 2.2 Sampling location

The study location was selected at the site of a USGS water quality platform located in LVB
(37°06'59.8", 114°50'51.4") near where treated WWTP effluent from the LVW enters Lake
Mead (Fig. 1). The water depth was 6.7 m during the sampling period (June 10 to July 9, 2008)
which was considerably less than the 18 m depth at the same location during a 2006 sampling
reported by Rosen et al. (2010) as water levels in Lake Mead have continued to drop over time.

113 2.3 Sampling methodology

114 The study design included the sampling of the water column and the sediment infiltrated water, 115 which required different sampling techniques. For both compartments, different configurations

of the SPMD and POCIS were used to measure the concentrations of dissolved SOCs. For the
water column measurements, SPMDs (460 cm² low density polyethylene tube containing 1.0 mL
of purified triolein) and POCIS (41 cm² polyethersulfone membrane disk containing 200 mg of
Oasis HLB) were constructed to meet the standard surface area to sorbent/lipid volume (SA/V)
specifications described by Huckins et al. (2006) and Alvarez et al. (2004, 2007). Stainless steel
deployment canisters, each containing four SPMDs or six POCIS, were suspended from the
water quality platform at depths of 3.0, 4.7, and 6.7 meters.

123

124 Sampling of the sediment infiltrated water required the design and construction of a custom 125 sediment sampling probe to bury the SPMDs and POCIS at depths of 0-10 cm, 10-20 cm, and 126 20-30 cm (Fig. 2). This design consisted of six perforated metal probes (3.8 cm in diameter, 30.5 127 cm long) attached to a 0.64 cm thick aluminum plate. The plate was attached to cables secured 128 to a buoy on the surface and had a location to attach weights as needed to aid in burying the probes into the sediment. Inside each of the metal probes, three mini-SPMDs (77 cm², 0.17 mL 129 triolein) or three mini-POCIS (20.5 cm², 100 mg Oasis HLB) were contained at specific 130 131 intervals. The mini-SPMDs and mini-POCIS conformed to the standard SA/V ratios previously 132 defined which allows for established uptake models to be applied when estimating water 133 concentrations (Goodbred et al., 2009). Racks constructed of Teflon spacers and a stainless steel 134 support rod held the mini-SPMDs and mini-POCIS in place while limiting the movement of 135 water between the three sampling chambers.

136

137 2.4 Processing and analysis

138 The passive samplers were processed and analyzed according to published procedures and 139 described in more detail in the Supplementary Materials (Alvarez et al., 2008a, 2008b; Jones-140 Lepp et al., 2004, 2010; Petty et al, 2000). Briefly, SPMDs were cleaned, dialyzed in hexane to 141 recover sequestered chemicals, filtered, and fractionated using a size exclusion chromatography 142 (SEC) system and gravity column reactive cleanup to isolate the chemicals of interest from other 143 sampled non-target chemicals. Following cleanup, SPMD dialysates designated for analysis of 144 PAHs and performance reference compounds (PRCs) were analyzed using an Agilent 6890 gas 145 chromatograph (GC, Agilent Technologies, Wilmington, DE) with a 5973N mass selective 146 detector (MSD, Agilent Technologies, Palo Alto, CA). Analyses of PCB, PBDE, and 147 chlorinated pesticide were performed using a Hewlett-Packard 5890 GC equipped with an 148 electron capture detector (ECD, Hewlett-Packard, Palo Alto, CA) following cleanup and 149 fractionation of the SPMD dialysates.

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151 The POCIS were cleaned followed by extracting the sampled chemicals from the POCIS sorbent 152 using 25 mL of a 80:20 (v:v) dichloromethane:methyl-*tert*-butyl ether solution for POCIS 153 designated for the analysis of OWCs and 40 mL of methanol for POCIS designated for the 154 analysis of pharmaceuticals or screening by the YES assay. The OWCs were analyzed using the 155 GC/MSD system previously described for the PAH analyses.

156

Pharmaceuticals and illicit drugs were analyzed by a Varian 500MS (Walnut Creek, CA) ion trap
mass spectrometer, configured with an electrospray ion source, and a Varian 212-liquid
chromatograph. An external standard calibration was used to nominally quantify the analytes.
Due to potentially interfering materials co-extracted with the pharmaceuticals and illicit drugs,

161 the analyses were performed using the collision induced dissociation (CID) mode for both 162 identification and for calculating the concentration of the analytes of interest. Two to three 163 product ions were used for identification and the most abundant product ion was chosen for 164 quantification.

165

SPMDs and POCIS were also screened for estrogenicity by the YES assay. None of the SPMDs in the sediment probe sampler were screened by the YES due to insufficient numbers of samplers available. Samples designated for the YES assay were solvent exchanged into histological grade alcohol and screened without any rigorous cleanup, with the exception of the SPMDs, to prevent removal of unknown but bioactive (estrogenic) chemicals. SPMD dialysates required SEC fractionation to remove any estrogenicity due to residual methyl oleate in the triolein used in the SPMD (Lebo et al., 2004; Rastall et al., 2004).

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174 All SPMDs designated for chemical analyses were fortified with PRCs (1.0 µg each of

acenaphthene- d_{10} , acenaphthalene- d_{10} , fluorine- d_{10} , phenanthrene- d_{10} , pyrene- d_{10} , and 20 ng each

176 of PCB congeners 14, 29, and 50) prior to use to provide a measure of the effect of

177 environmental conditions had on the sampler's performance. Dibenz[a,h]anthracene- d_{14} (1.0 µg)

178 was also added to each SPMD to serve as a photolysis surrogate to indicate if photolysis of

179 sequestered PAHs had occurred (Table S10).

180

181 Quality control measures used in this study included field and laboratory blanks, surrogate

182 recovery spikes of target chemicals in both the SPMD and POCIS matrices, multi-point

183 calibration curves with the instrumental analyses, and positive and negative controls for the YES

assay. Mean recoveries of the test chemicals were 71%±40% (Tables S11-S12). The blanks had
few chemicals present at detectable levels. With the exception of diethylhexylphthalate and
cholesterol, most blank detections were below 20 ng/sample (Tables S13-S15).

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188 **3. Results and Discussion**

189 In the current study, it was difficult to determine a clear gradient as the lake depth at this location 190 had dropped to 6.7 m from the 18 m depth two years prior by Rosen et al. (2010). Temperature 191 and specific conductance readings taken by the remote water quality station at least once per day 192 at depths of 1.0, 3.0, and 5.0 m showed no significant variation in measurements between each 193 depth. Temperature readings ranged from an average of $27.5^{\circ}C\pm 2.2$ to $26.5^{\circ}C\pm 2.0$ and the 194 specific conductance ranged from 1669 μ S/cm \pm 104 to 1718 μ S/cm \pm 100 for the 1.0 and 5.0 m 195 depths respectively. Rosen et al. (2010) determined that the SOCs were confined to a 6 m region 196 of the water column nearest the lake bottom. As the total depth of the water column was 6.7 m 197 in the current study, mixing within the water column appears to be occurring resulting in less 198 defined gradient of SOCs and water quality parameters.

199

Most of the chemicals which are commonly associated with WWTP effluents and have greater water solubilities were measured at elevated concentrations in the upper water column (Tables 1-2, S7, S9). Chemicals commonly associated with WWTP effluents, including the fragrance galaxolide (HHCB), the phosphate-based flame retardants, and the surfactant *p-tert*-octylphenol were all measured at the greatest concentrations in the upper water column. HHCB was present at 48 ng/L near the lake surface compared to a previous study reporting HHCB levels in LVW at 33 to 44 ng/L during June and July of 2004 (Osemwengie and Gerstenberger, 2004). In the same

207 study, they measured HHCB at 0.12 and 0.22 ng/L in Lake Mead, but at a location downstream 208 of LVB which would have resulted in considerable dilution of any effluents entering Lake Mead 209 from LVW. Reiner and Kannan (2011) reported HHCB concentrations ranging from 3.95 to 210 25.8 ng/L in the Hudson River which received treated effluents from numerous communities. 211 Hydrocodone (120 ng/L), a narcotic analgesic and cough suppressant, and the illicit drug 212 methamphetamine (12 ng/L) were detected only in the upper water column. The estimated 213 concentrations for these drugs are considered nominal values as no surrogate standards were 214 used in the LC/MS analyses. Although a true measure of ion suppression of the pharmaceuticals 215 and illicit drugs could not be determined, the effect due to co-extracted chemicals is expected to 216 be minimal as large potentially interfering chemicals are not sampled by the POCIS. Jones-Lepp 217 et al. (2004) previously measured methamphetamine in LVW (referred to as an unnamed site in 218 Nevada in the publication) at concentrations of 1.3 and 0.8 ng/L during the summer and winter 219 months of 2002 and 2003, respectively. Clindamycin, a lincosamide antibiotic widely used as a 220 topical treatment for acne, was measured at trace concentrations at the 3.0 m depth and at 37 221 ng/L at the 6.7 m depth. Phenol and *para*-cresol, chemicals which can originate from multiple 222 sources including WWTP effluents, were only present at the bottom of the water column.

223

The more hydrophobic chemicals such as PAHs were present at the highest concentrations at the lake bottom and in the upper 20 cm of the sediment infiltrated water (Tables 1, S3-S4). The distribution of PAHs moves from the water column for PAHs with log octanol-water partition coefficients (K_{ow} s) <5.5 (naphthalene to pyrene) to the sediment infiltrated water for PAHs with log K_{ow} s>5.5 (benz[*a*]anthracene to benzo[*g*,*h*,*i*]perylene). Rosen et al. (2010) reported finding the greatest concentrations of PAHs in the upper layer (4 m) of LVB water; however, that study

focused solely on PAHs and substituted PAHs ranging from naphthalene to pyrene excluding the ones with higher log K_{ow} s. The data from this study are consistent with previous studies where the lower molecular weight, lower K_{ow} , PAHs are present near the surface of the lake (Lico and Johnson, 2007; Rosen et al., 2010). Many of the PAHs detected are characteristic of petrogenic sources and may be present from recreational boat use in the area as suggested by Lico and Johnson (2007).

236

237 Few chlorinated pesticides and PCBs were identified in either the water column or sediment 238 infiltrated water with little variation in their water concentrations at any sampling depth (Tables 239 1, S5-S6). Endosulfan and its degradation product, endosulfan-II, were found at the greatest 240 concentrations at the 3.0 and 4.7 m depths in the water column. Endosulfan is an acutely toxic 241 insecticide reported as a known endocrine disruptor (Soto et al., 1994; Crisp et al., 1998). The 242 former BMI Complex in southeastern Las Vegas Valley housed different pesticide 243 manufacturers, some of which are suspected to have produced endosulfan (Sahu, 2006). The 244 BMI Complex historically dumped wastes into LVW leading to the direct input of numerous 245 contaminants into Lake Mead. Endosulfan has also been shown to be amenable to long distance 246 atmospheric transport leading to measurable concentrations of the pesticide in water bodies 247 hundreds of kilometers away from the location of initial use (LeNoir et al., 1999; Muir et al., 248 2004). Concentrations of endosulfan at LVB were within a factor of 2 or 3 to reported 249 concentrations in the Sierra Nevada mountain lakes due to atmospheric deposition (LeNoir et al., 250 1999).

251

252 The chemical found at the greatest concentrations in the upper portions of the water column, was 253 the industrial plasticizer N-butyl benzenesulfonamide (NBBS) at concentrations of 1600 ng/L 254 and 1100 ng/L at the 3.0 and 4.9 m depths, respectively (Tables 2, S7). NBBS is a precursor in 255 the production of nylon and other polyamide polymers (Strong et al., 1991). NBBS is a 256 neurotoxin reported to cause dose-dependent motor dysfunction in rabbits and rats but is rapidly 257 eliminated from the body requiring a prolonged exposure to high concentrations (>1 mg/kg) to 258 produce a neurotoxic effect (Strong et al., 1991; Kumar et al., 2007). No experimental data 259 exists on the acute, reproductive, or developmental toxicity of NBBS to fish; however, the US 260 Environmental Protection Agency used predictive models to estimate that the risk to aquatic 261 species was relatively low (US EPA, 2003). With its low log K_{ow} of 2.17, NBBS can be found in 262 the water column and tends not to bioaccumulate in aquatic organisms (US EPA, 2003; Kumar et 263 al., 2007). It is unknown if any polymer manufacturers using NBBS were located within the 264 LVW/LVB drainage area.

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266 Phenol, *para*-cresol, and indole were at the highest concentrations in the top 0-10 cm of the 267 sediment and also greatest in the water column at the 6.7 m (lake bottom) depth possibly 268 indicating that the sediment is acting as a contaminant source to the overlying water (Table 2, 269 Fig. 3). Para-cresol was previously measured in sediments collected from LVB in 1992, 1995, 270 and 1998 (Bevans et al., 1996, Covay and Beck, 2001). Phenol was also measured in the 271 sediment collected in 1998. A source reconnaissance study in Ohio found para-cresol present in 272 100% of sediment samples and 67% of water samples regardless of their spatial relation to 273 WWTPs (Tertuliani et al., 2008). This along with evidence suggesting that the LVW plume is 274 moving across the surface of the lake suggests that WWTP effluents from LVW are not the sole

source of all chemicals in LVB as surface runoff, shallow ground water, atmospheric inputs, and
irrigation are all potential inputs into Lake Mead (Rosen et al., 2010).

277

278 A measurable level of estrogenicity from sequestered SOCs was present in POCIS extracts at 279 each sampling depth in the water column and the sediment infiltrated water. In the water 280 column, the estimated estrogenicity was 5 to 9-fold greater at the 3.0 and 4.7 m depths which 281 correlates with the greater occurrence of many SOCs related to WWTP effluents at these depths 282 (Table 3). Although not directly measured in this study, it is assumed that many of the steroidal 283 hormones originating in WWTP effluents would follow the effluent plume. From the SPMDs, 284 an estrogenic response was detected in only the 6.7 m sampler. Since the SPMD samples only 285 nonpolar, nonionic organic chemicals, it is not surprising that the greatest estrogenic response 286 was at the sediment-water interface where hydrophobic chemicals were at the greatest 287 concentrations.

288

Higher concentrations of some SOCs in the upper layer of the sediment infiltrated water and at the sediment-water interface suggests that the sediment may be acting as a secondary source for some chemicals. This could be a concern to the ecosystem health as SOCs associated with the sediment could impact bottom-dwelling organisms and have repercussions throughout the food chain. Water quality criteria and toxicity data is limited for many of the chemicals identified at the sediment-water interface. What data was available suggested that the concentrations measured in this study were significantly below any acute toxicity levels.

296

297 The design of the sediment probe device provided a means of successively deploying SPMDs 298 and POCIS at fixed depths in the sediment. The concept of using perforated tubes containing the 299 sampling media has been explored before by De Jonge and Rothenberg (2005) who used 300 perforated tubes containing a solid adsorbent material to measure the flux of PAHs and 301 pesticides in soils. The Peepers sampling device for infiltrated water measurements also has a 302 similar design with the exception that a diffusion membrane is in direct contact with the 303 sediment (Hesslein, 1976). In our study, only minimal sediment was found in each sampling 304 chamber indicating free exchange of water between the passive samplers and the surrounding 305 sediment. Comparing the PRC loss rates (Table S10) from the SPMDs in the sediment probes to 306 that of the water column samplers, it can be qualitatively determined that water exchange 307 between the water in the sampling chambers and the infiltrated water in the sediment did occur, 308 albeit at a reduced rate.

309

The use of a sediment probe device as described in this work is advantageous only when the bed sediment is relatively soft allowing the probes to bury themselves. This system would not work in rocky areas. Some disruption of the bed sediment will likely occur as the probes are inserted, however, effects from this disruption should be minimized as the samplers are providing a measure of the average chemical composition of the infiltrated water over weeks to months.

315

316 **4. Conclusions**

317 Lake sediment is a sink for many SOCs; however, partitioning between the sediment and the 318 overlying water could result in the sediment acting as a secondary contaminant source. Using a 319 combination of passive samplers in the water column as well as a custom sediment probe device

to bury passive samplers in the sediment, it was determined that the water at the sediment-water interface had a distinct profile from the rest of the water column. Measurement of the direct risk to sediment-dwelling organisms was beyond the scope of this project, but there is evidence to suggest that the whole water column should be considered when determining the health of a body of water.

325

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483 Figure 1. Sampling location in Las Vegas Bay, Lake Mead National Recreation Area, Nevada.

- 484 The markers labeled 1, 2, and 3 refer to the City of Las Vegas wastewater treatment plant
- 485 (WWTP), the Clark County WWTP, and the Henderson WWTP, respectively.

486 487

- 488 Figure 2. Diagram of the sediment probe device. The device contained six probes, each capable
- 489 of holding three POCIS or three SPMDs at depth intervals of 0-10, 10-20, and 20-30 cm.
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- 491
- 492 Figure 3. Distribution of total organic wastewater chemicals measured by polar organic chemical
- 493 integrative samplers (POCIS) suspended in the water column and buried in sediment. The
- 494 number above each bar is the total concentration (ng/L) of chemicals.

1 Supplementary Material

2

Bottom sediment as a source of organic contaminants in Lake Mead, USA

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- 90 sediment in Las Vegas Bay, Lake Mead, NV.
- 91

92 Table S15. Concentrations of waste indicator chemicals in polar organic chemical integrative

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column and buried in the sediment in Las Vegas Bay, Lake Mead, NV.

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98 1. Processing and analysis of passive samplers

- The passive samplers were processed and analyzed according to published procedures
 (Alvarez et al., 2008a, 2008b; Jones-Lepp et al., 2004, 2010; Petty et al, 2000).
- 101

1.1 SPMDs – Briefly, SPMDs were cleaned to remove any films or salts on the membrane 102 surfaces and then the sequestered chemicals were recovered using a standard two-stage 103 104 dialysis with hexane. Following dialysis, the dialysates were concentrated and filtered prior to fractionation on a size exclusion chromatography (SEC) system to isolate the chemicals of 105 interest from other sampled non-target chemicals. Following SEC, dialysates designated for 106 107 PAH analyses were applied to a tri-adsorbent column consisting of phosphoric acid silica gel, potassium hydroxide impregnated silica gel, and silica gel to clean-up the dialysates prior to 108 analysis. The enriched samples were then analyzed for PAHs and performance reference 109 compounds (PRCs) using an Agilent 6890 gas chromatograph (GC, Agilent Technologies, 110 Wilmington, DE) with a 5973N mass selective detector (MSD, Agilent Technologies, Palo 111 Alto, CA) with an HP-5MS (30 m, 0.25 mm ID, 0.25 um film thickness) capillary column 112 (Agilent Technologies, Wilmington, DE). Samples were at a volume of 1 mL with 1 µL 113 injected using a cool-on-column inlet.

114 115

SPMDs designated for PCB, PBDE, and chlorinated pesticide analyses underwent additional
cleanup following SEC by applying the samples to Florisil columns followed by fractionation
on silica gel columns. The PCB and chlorinated pesticide/PBDE fractions generated from
the silica gel step were analyzed by a Hewlett-Packard 5890 GC equipped with an electron
capture detector (ECD, Hewlett-Packard, Palo Alto, CA) and a DB-35MS (30 m, 0.25 mm
ID, 0.25 um film thickness) capillary column (J&W Scientific, Folsom, CA). Samples were
at a volume of 1 mL with 1 μL injected using a cool-on-column inlet.

123

1.2 POCIS - The POCIS were gently rinsed with deionized (DI) water to remove any loose 124 materials on the surface of the samplers prior to extractions. Each POCIS was individually 125 extracted by transferring the sorbent from inside the POCIS with DI water into a pre-cleaned 126 empty 25 mL solid-phase extraction (SPE) tube fitted with a polyethylene frit. A second frit 127 was placed on top of the sorbent bed and then the residual water was removed by pulling air 128 through the cartridges for approximately 10 minutes. To recover the sampled chemicals from 129 the POCIS sorbent, 25 mL of a 80:20 (v:v) dichloromethane:methyl-tert-butyl ether solution 130 was used for POCIS designated for the analysis of OWCs and 40 mL of methanol was used 131 for POCIS designated for the analysis of pharmaceuticals or screening by the YES assay. 132 Following extraction, the samples were concentrated using a rotary evaporation system and 133 high purity nitrogen blowdown, filtered, and transferred into autosampler vials for analysis. 134 The wastewater indicator chemicals were analyzed using the GC/MSD system previously 135 described for the PAH analyses. 136

- Pharmaceuticals and illicit drugs were analyzed by a Varian 500MS (Walnut Creek, CA) ion trap mass spectrometer, configured with an electrospray ion source, and a Varian 212-liquid chromatograph, was used for all analyses. Mid-range calibration standards (0.5 and 1 ng/ μ L) were analyzed at the beginning and end of each analytical day. A volume of 5 μ L was injected for each standard. Linearity and precision of the daily calibration standards were measured from an initial 3-pt calibration curve prepared and analyzed weekly. A volume of 10 μ L was injected for each sample extract. An external standard calibration, along with POCIS rate uptake information, was used to nominally quantify the analytes.
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The 500MS was run in the positive ionization mode under the following conditions: ES
needle, 5 kV; drying gas, 20 psi and 350° C; housing chamber, 50° C; nebulizer gas, 40 psi;
spray shield, 600 V; capillary voltages were set dependent upon the optimized response of
the product ions of interest.

- 151 Due to potentially interfering materials co-extracted with the pharmaceuticals and illicit 152 drugs, the analyses were performed using the collision induced dissociation (CID) mode for 153 both identification and for calculating the concentration of the analytes of interest. Two to 154 three product ions were used for identification and the most abundant product ion was chosen 155 for quantification. The precursor ion and most abundant product ion that were used to 156 identify and quantify the pharmaceuticals and illicit drugs, and their limits-of-detection 157 (LOD, on-column) are shown in Table S2. Liquid chromatographic separations were 158 performed using an Ascentis Express C18 (Supelco-Aldrich, Bellefonte, PA) 2.7 µm particle 159 size, 3 cm x 2.1 mm column, coupled with a Varian guard column (MetaGuard 2.0 mm 160 Pursuit XRs 3µm C18). The flow rate through the column was 200 µL/min, with the 161 following gradient elution conditions: initial conditions mobile phase A 100%, hold for 2 162 min; 3 min gradient to 30% A:70% B, hold for 5 min; 3 min gradient to 100% A, hold for 2 163 min; end run, 5 min equilibration time between analyses. Compositions of the mobile phases 164 were as follows: (A) deionized water/0.5% formic acid, and (B): 82% methanol/18% 165 acetonitrile/0.5% formic acid. 166
- 167

168 *1.3 Estrogenicity Screening* - POCIS designated for the YES assay were solvent exchanged into
1 mL of histological grade alcohol and 100 µL of each sample was serially diluted and
170 screened without any rigorous cleanup to prevent removal of unknown but bioactive
171 (estrogenic) chemicals. The YES assay uses a yeast cell transfected with an estrogen
172 receptor. Upon binding with estrogen or an estrogen mimic, a series of reactions occurs
173 resulting in a color change which can be measured spectrophotometrically (Rastall et al.,
174 2004; Alvarez et al., 2008a).

SPMDs deployed in the water column were also screened for estrogenicity by the YES assay.
Prior to screening with the YES assay, SPMD dialysates required SEC fractionation to
remove any estrogenicity due to residual methyl oleate in the triolein used in the SPMD
(Lebo et al., 2004). Following SEC, the SPMD fractions were solvent exchanged into 1 mL
of histological grade alcohol and 100 µL of each sample was serially diluted on the YES
plate. None of the SPMDs in the sediment probe sampler were screened by the YES due to
insufficient numbers of samplers available.

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184 2. Estimation of time-weighted average water concentrations

The estimation of time-weighted average water concentrations of chemicals sequestered by the SPMD and POCIS requires knowledge of the sampling rate for each chemical along with the sampling duration. Using models previously developed (Alvarez et al., 2007; Huckins et al., 2006), data from the analysis of the PRCs added to the SPMDs, and experimentallyderived or theoretically-estimated sampling rates, the bioavailable (i.e., via respiration from the dissolved phase) concentrations of SOCs in the SPMD and POCIS can be estimated.

- PRCs are analytically non-interfering organic compounds with moderate to high fugacity 192 from SPMDs that are added to the lipid prior to membrane enclosure and field deployment 193 (Huckins et al., 2006). By applying the amount of PRCs lost during field exposures to a 194 third-order regression model, a site-specific sampling rate can be determined to increase the 195 accuracy of the water concentration estimates. The models describing the uptake and the use 196 of online calculators to calculate water concentrations from SPMD data have been discussed 197 in detail (Alvarez et al., 2010; Huckins et al., 2006). Due to the strong sorptive properties of 198 the adsorbents used in the POCIS, attempts to incorporate PRCs to cover a wide range of 199 analytes into the POCIS have failed (Alvarez et al., 2007). Without PRCs, the estimation of 200 water concentrations from POCIS data requires availability of sampling rates, potentially 201 limiting the number of compounds which can be determined (Alvarez et al., 2007; 2010). 202
- 204Uptake of chemicals into passive samplers generally follows linear, curvilinear and205equilibrium phases of sampling. Integrative (or linear) sampling is the predominant phase for206compounds with log K_{OW} values ≥ 5.0 and exposure periods of up to one month in SPMDs207and for most of the chemicals tested in the POCIS. During the linear uptake phase, the208ambient chemical concentration (C_w) is determined by
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210 211 $C_{\rm w} = N/R_{\rm s}t \tag{1}$

where *N* is the amount of the chemical accumulated by the sampler (typically ng), R_s is the sampling rate (L/d), and *t* is the exposure time (d). Previous data indicates that many chemicals of interest sampled by the POCIS remain in the linear phase of sampling for at least 56 d (Alvarez et al., 2007; 2010), therefore, the use of a linear uptake model (Eqn. 1) for the calculation of ambient water concentrations was justified.

For the POCIS, the availability of R_s values can limit which chemicals it is possible to estimate water concentrations. For this work, a series of R_s values were used which originated from published and unpublished sources (Alvarez, unpublished work; Alvarez et al., 2007; Bartlet-Hunt et al., 2011; Harman et al., 2008; MacLeod et al., 2007). Water concentrations were performed only for chemicals which had at least one detection.

For SPMDs, regression models have been created which estimate a chemical's site specific *R*_s and its C_w based on the log K_{OW} of the chemical, the PRCs release rate constant (k_e) and SPMD-water partition coefficient (K_{sw}) (Huckins et al., 2006). A PRCs k_e is determined from the amount of PRC initially added to the SPMD (N_o) and the amount remaining (N) as shown in Equation 2. The log K_{sw} is determined from a regression model of the PRCs log K_{OW} as shown in Equation 3 where a_0 is the intercept determined to be -2.61 for PCBs,

as shown in Equation 4 where V_s is the volume of the SPMD. 231 232 233 $k_{\rm e} = - [\ln(N/N_{\rm o})]/t$ (2) 234 $\log K_{\rm sw} = a_0 + 2.321 \log K_{\rm ow} - 0.1618 (\log K_{\rm ow})^2$ 235 (3) 236 $R_{s-PRC} = V_s K_{sw} k_e$ 237 (4) 238 The extrapolation of C_w from measured values of N requires knowledge of a chemical's site-239 specific sampling rate (R_{si}) which is determined from a third-order polynomial (Eqn. 5) 240 where $\alpha_{(i/PRC)}$ is the compound-specific effect on the sampling rate and the relationship 241 between the R_{s-PRC} and R_{si} (Eqn. 6). 242 243 $\log \alpha_{(i/PRC)} = 0.0130 \log K_{OW}^{3} - 0.3173 \log K_{OW}^{2} + 2.244 \log K_{OW}$ (5) 244 245 $R_{\rm si} = R_{\rm s-PRC} (\alpha_{\rm i} / \alpha_{\rm PRC})$ (6) 246 247 248 The C_w of a chemical in the water can then be calculated by 249 $C_{\rm w} = N/(V_{\rm s}K_{\rm sw}[1-\exp(-R_{\rm s}t/V_{\rm s}K_{\rm sw})])$ 250 (7)251 252 Rates of diffusion of SOCs between the sediment and a passive sampler in the sediment 253 probe device are controlled by a number of variables including the organic carbon content of 254 255 the sediment, the log K_{ow} of the chemical, the length of the diffusion pathway, and the hydraulic permeability of the sediment (Huckins et al., 2006). Most of these variables are 256 difficult to measure in situ and may require complicated transport models to describe the 257 258 uptake into SPMDs. For this work, the concentration gradient in relation to the sediment depth was of greater importance than the actual infiltrated water concentration. Therefore, 259 two assumptions were made to simplify the estimation of the ambient water concentrations: 260 261 1) PRC losses would adjust the chemical sampling rates to account for water movement and resistance to chemical transport; and 2) using standard first-order uptake models along with 262 263 the PRCs would provide a reasonable approximation of the chemical concentrations in the infiltrated water. 264 265 3. Method detection limits and method quantitation limits 266 The method detection limit (MDL) and method quantification limit (MQL) for analysis of 267 POCIS and SPMD samples were determined for each analyte by measuring the values of 268 coincident instrumental chromatographic peaks in all field blank samples for each analyte 269 (Tables S3-S8). Determination of MDL and MQL values have been described by Keith 270 271 (1991). The MDL was operationally defined as the mean of field blanks plus three standard 272 deviations. The MQL was operationally defined as the mean of field blanks plus ten-standard deviations. For individual analytes having no coincident chromatographic peak, an assumed 273

PAHs, nonpolar pesticides and -3.20 for polar pesticides. The R_{s-PRC} can then be calculated

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value equal to the low sample reject for the instrumental method (operationally defined as

- 275 20% of the concentration of the lowest standard concentration used for the calibration curve)
- was used to calculate the mean. In the cases where the calculated values of the MQLs were
- below the level of the calibration curve employed in the analysis, the MQLs were set at the
- value of the lowest level of the calibration curve employed in quantifying concentrations ofan analyte.
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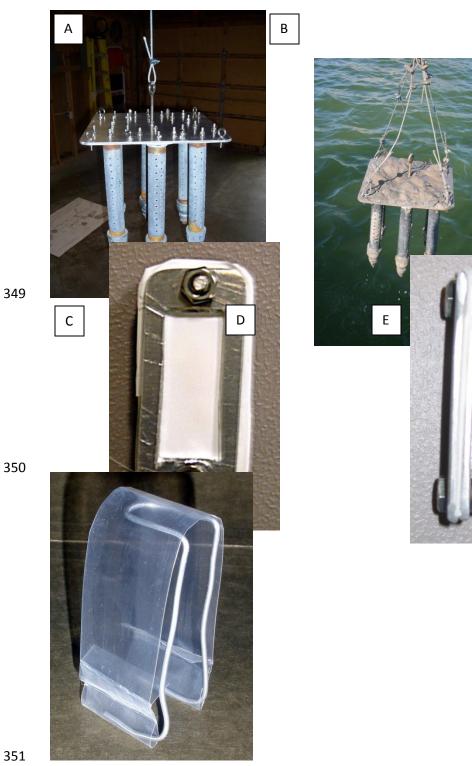
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- 341 semipermeable membrane devices (SPMDs), bioassays and chemical analysis. Environ Sci
- 342 Pollut Res 11, 240-253.
- 343

- Figure S1. Photographs of the sediment probe device and the miniature semipermeable
- membrane devices (SPMDs) and polar organic chemical integrative samplers (POCIS) used in
- the sediment probes. A sediment probe device; B sediment probe device during retrieval; C
- 347 mini-POCIS unit; D side view of two mini-POCIS units; E mini-SPMD.



Waste Indie	cator Chemicals ^a	Polycyclic aromatic hydrocarbons ^b	Chlorinated Pesticides / PCBs / PBDEs ^b
Tetrachloroethylene	Anthracene	Naphthalene	Trifluralin
Bromoform	Diazinon	Acenaphthylene	Hexachlorobenzene (HCB)
Isopropylbenzene	Musk Ambrette	Acenaphthene	Pentachloroanisole (PCA)
(cumene)		-	
Phenol	Carbazole	Fluorene	Tefluthrin
1,4-Dichlorobenzene	Caffeine	Phenanthrene	alpha-Benzenehexachloride (a-BHC)
d-Limonene	Traseolide (ATII)	Anthracene	Diazinon
Acetophenone	Galaxolide (HHCB)	Fluoranthene	Lindane
para-Cresol	Tonalide (AHTN)	Pyrene	beta-Benzenehexachloride (b-BHC)
Isophorone	Musk Xylene	Benz[a]anthracene	Heptachlor
Camphor	Carbaryl	Chrysene	delta-Benzenehexachloride (d-BHC)
Menthol	Metalaxyl	Benzo[b]fluoranthene	Dacthal
Naphthalene	Bromacil	Benzo[k]fluoranthene	Chlorpyrifos
Methyl salicylate	Anthraquinone	Benzo[a]pyrene	Oxychlordane
Dichlorvos	Musk Ketone	Indeno[1,2,3- <i>c</i> , <i>d</i>]pyrene	Heptachlor Epoxide
Isoquinoline	Chlorpyrifos	Dibenz[<i>a</i> , <i>h</i>]anthracene	trans-Chlordane
Indole	Fluoranthene	Benzo[g,h,i]perylene	trans-Nonachlor
2-Methyl naphthalene	Pyrene	Benzo[b]thiophene	o,p'-DDE
1-Methyl naphthalene	Tri(dichloroisopropyl) phosphate	2-methylnaphthalene	cis-Chlordane
2,6-Dimethylnaphthalene	Tri(butoxyethyl) phosphate	1-methylnaphthalene	Endosulfan
Cashmeran (DPMI)	Triphenyl phosphate	Biphenyl	p,p'-DDE
N,N-diethyltoluamide (DEET)	Diethylhexylphthalate (DEHP)	1-ethylnaphthalene	Dieldrin
Diethyl phthalate	Benzo[a]pyrene	1,2-dimethylnaphthalene	o,p'-DDD
p-tert-Octylphenol	Cholesterol	4-methylbiphenyl	Endrin
Benzophenone		2,3,5-trimethylnaphthalene	cis-Nonachlor
Tributyl phosphate	Pharmaceuticals ^a	1-methylfluorene	o,p'-DDT
Ethyl citrate	Azithromycin	Dibenzothiophene	p,p'-DDD
Cotinine	Clarithromycin	2-methylphenanthrene	Endosulfan-II
Celestolide (ADBI)	Clindamycin	9-methylanthracene	p,p'-DDT
Prometon	Hydrocodone	3,6-dimethylphenanthrene	Endosulfan Sulfate
Atrazine	MDMA (Ecstasy)	2-methylfluoranthene	p,p'-Methoxychlor
Phantolide (AHMI)	Methamphetamine	Benzo[b]naphtho[2,1-d]thiophene	Mirex
4-Octylphenol	N,N-DMPEA	Benzo[e]pyrene	cis-Permethrin
Tri(2-chloroethyl) phosphate	Pseudoephredrine	Perylene	trans-Permethrin
N-butyl benzenesulfonamide	Roxithromycin	3-methylcholanthrene	Total PCBs
Phenanthrene	Urobilin		PBDE congeners 28, 27, 99, 100, and 153

Table S1. Chemicals selected for analysis in passive samplers deployed in Lake Mead.

^a Chemicals under this general chemical classification were measured in polar organic chemical integrative sampler

355

(POCIS) extracts. ^b Chemicals under this general chemical classification were measured in semipermeable membrane device (SPMD)

extracts.

Table S2. Monitoring ions and limits of detection (LOD) used for the LC/MS/MS analyses of

360 pharmaceuticals and illicit	drugs.
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Compound	Precursor ion	Product ion	LOD ^a
Urobilin hydrochloride	$591.3 (M + H - HCl)^+$	$343.3 [M+H-HCl - 2(C_7H_{10}NO)]^+$	0.4
Azithromycin	749.5 $(M+H)^+$	591.4 $(M+H-C_8H_{16}O_2N)^+$	0.5
Roxithromycin	$859.5 (M+Na)^+$	$755.4 (M+Na-C_4H_9O_3)^+$	1
Clarithromycin	$748.4 (M+H)^+$	590.1 $(M+H-C_8H_{16}O_2N)^+$	1
Clindamycin	$425.2 (M+H)^+$	$377.2 (M+H-SH-CH_3)^+$	1
Methamphetamine	$150 (M+H)^+$	$119 (M+H-CH_3NH_2)^+$	2
MDMA(Ecstasy)	$194 (M+H)^+$	$163.0 (M-CH_3NH_2+H)^+$	1
Pseudoephedrine	$166 (M+H)^+$	$148.2 (M+H-H_2O)^+$	3
Hydrocodone	$300 (M+H)^+$	199 (M+H-C ₅ H ₁₁ NO)+	2

^a as determined using MacDougall et al., guidelines (MacDougall and Crummett 1980), ng on-column.

Table S3. Water concentrations of polycyclic aromatic hydrocarbons (PAHs) estimated from

residues in semipermeable membrane devices deployed in the water column in Las Vegas Bay,

367 Lake Mead, NV.

		Sampler			r Depth in Water Column		
	MDL	MQL	3.0 m	4.7 m	6.7 m		
	pg/L	pg/L	pg/L	pg/L	pg/L		
Naphthalene	140	680	680	680	1400		
Acenaphthylene	28	140	<28	<28	<28		
Acenaphthene	20	100	<20	<20	110		
Fluorene	14	72	74	73	400		
Phenanthrene	190	440	260	250	420		
Anthracene	11	53	56	55	63		
Fluoranthene	48	140	330	380	320		
Pyrene	4.3	22	290	360	410		
Benz[<i>a</i>]anthracene	3.7	19	24	21	34		
Chrysene	3.8	19	140	150	160		
Benzo[b]fluoranthene	3.7	19	47	43	66		
Benzo[k]fluoranthene	3.9	19	51	45	36		
Benzo[a]pyrene	4.1	20	27	24	77		
Indeno[1,2,3- <i>c</i> , <i>d</i>]pyrene	4.8	24	65	29	94		
Dibenz[<i>a</i> , <i>h</i>]anthracene	4.3	22	<4.3	<4.3	<4.3		
Benzo[g,h,i]perylene	5.2	26	71	62	100		
Benzo[b]thiophene	530	2600	<530	<530	<530		
2-methylnaphthalene	560	1500	<560	<560	940		
1-methylnaphthalene	47	230	230	230	700		
Biphenyl	42	210	<42	210	210		
1-ethylnaphthalene	14	71	<14	<14	160		
1,2-dimethylnaphthalene	18	92	<18	<18	300		
4-methylbiphenyl	17	85	<17	<17	<17		
2,3,5-trimethylnaphthalene	6.2	31	70	99	340		
1-methylfluorene	97	210	130	120	280		
Dibenzothiophene	14	72	<14	73	160		
2-methylphenanthrene	6.3	31	70	67	43		
9-methylanthracene	5.1	26	<5.1	<5.1	<5.1		
3,6-dimethylphenanthrene	4.0	20	50	45	33		
2-methylfluoranthene	3.9	20	25	22	33		
Benzo[b]naphtho[2,1-d]thiophene	4.2	21	<4.2	<4.2	<4.2		
Benzo[e]pyrene	4.1	21	110	120	200		
Perylene	3.8	19	75	89	140		
3-methylcholanthrene	5.9	30	<5.9	<5.9	<5.9		

368 MDL – method detection limit; MQL – method quantitation limit

Less than (<) values are results below the method detection limit.

370 Results in *italic* type are estimated values greater than the method detection limit but less than the method

371 quantitation limit. These values have a greater amount of uncertainty in the absolute value.

372 Results in **bold** type are reportable values greater than the method quantitation limit.

Table S4. Water concentrations of polycyclic aromatic hydrocarbons (PAHs) estimated from

residues in semipermeable membrane devices buried in the sediment in Las Vegas Bay, Lake

376 Mead, NV.

			Sampl	er Depth in S	ediment
	MDL	MQL	0-10 cm	10-20 cm	20-30 cm
	pg/L	pg/L	pg/L	pg/L	pg/L
Naphthalene	11000	28000	<11000	<11000	<11000
Acenaphthylene	180	910	<180	<180	<180
Acenaphthene	140	670	<140	<140	<140
Fluorene	99	500	510	500	490
Phenanthrene	430	1300	450	440	<430
Anthracene	76	380	<76	<76	<76
Fluoranthene	410	1200	<410	<410	<410
Pyrene	38	190	220	200	190
Benz[a]anthracene	36	180	210	190	170
Chrysene	36	180	210	180	170
Benzo[b]fluoranthene	35	180	210	180	<35
Benzo[k]fluoranthene	39	190	230	200	190
Benzo[a]pyrene	41	200	240	<41	<41
Indeno[1,2,3- <i>c</i> , <i>d</i>]pyrene	49	250	290	<49	<49
Dibenz[<i>a</i> , <i>h</i>]anthracene	44	220	260	<44	<44
Benzo[g,h,i]perylene	54	270	320	280	<54
Benzo[b]thiophene	3400	17000	<3400	<3400	<3400
2-methylnaphthalene	3200	9300	<3200	<3200	<3200
1-methylnaphthalene	300	1500	1500	<300	<300
Biphenyl	280	1400	<280	<280	<280
1-ethylnaphthalene	97	490	<97	<97	<97
1,2-dimethylnaphthalene	120	620	<120	<120	<120
4-methylbiphenyl	120	570	<120	<120	<120
2,3,5-trimethylnaphthalene	49	250	<49	<49	<49
1-methylfluorene	490	1400	<490	<490	<490
Dibenzothiophene	99	500	<99	<99	<99
2-methylphenanthrene	50	250	<50	<50	<50
9-methylanthracene	43	210	<43	<43	<43
3,6-dimethylphenanthrene	36	180	<36	<36	<36
2-methylfluoranthene	36	180	<36	<36	<36
Benzo[b]naphtho[2,1-d]thiophene	37	190	<37	<37	<37
Benzo[e]pyrene	42	210	<42	<42	<42
Perylene	38	190	<38	<38	<38
3-methylcholanthrene	61	310	<61	<61	<61

377 MDL – method detection limit; MQL – method quantitation limit

378 Less than (<) values are results below the method detection limit.

379 Results in *italic* type are estimated values greater than the method detection limit but less than the method

380 quantitation limit. These values have a greater amount of uncertainty in the absolute value.

381 Results in **bold** type are reportable values greater than the method quantitation limit.

Table S5. Water concentrations of chlorinated pesticides, polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) estimated from residues in semipermeable membrane 383

384	polybrominated	l diphenyl ethers	(PBDEs) estimated	d from residues in semipermeable membrane	
			1 1 7 77		

385	devices deployed in the water column in Las Vegas	Bay, Lake Mead, NV.
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			Sampler Depth in Water Colu			
	MDL	MQL	3.0 m	4.7 m	6.7 m	
	pg/L	pg/L	pg/L	pg/L	pg/L	
Trifluralin	12	16	25	15	13	
Hexachlorobenzene (HCB)	0.89	2.2	25	26	13	
Pentachloroanisole (PCA)	19	52	<19	<19	<19	
Tefluthrin	64	150	<64	<64	<64	
alpha-Benzenehexachloride (a-BHC)	150	200	<150	<150	<150	
Diazinon	16000	20000	<1600	<1600	<1600	
Lindane	240	270	<240	<240	<240	
beta-Benzenehexachloride (b-BHC)	60	110	180	220	110	
Heptachlor	10	26	<10	<10	<10	
delta-Benzenehexachloride (d-BHC)	86	250	<86	<86	<86	
Dacthal	17	45	<17	<17	<17	
Chlorpyrifos	28	74	<28	<28	<28	
Oxychlordane	0.39	2.0	< 0.4	< 0.4	< 0.4	
Heptachlor Epoxide	28	74	<48	<48	<48	
trans-Chlordane	3.0	7.6	4.5	5.8	5.5	
trans-Nonachlor	5.1	12	<5.1	<5.1	<5.1	
o,p'-DDE	9.3	24	62	94	65	
cis-Chlordane	0.43	2.1	14	19	13	
Endosulfan	22	110	61	51	<22	
p,p'-DDE	16	27	100	140	160	
Dieldrin	20	53	28	40	<20	
o,p'-DDD	12	33	35	39	21	
Endrin	10	29	<10	<10	<10	
cis-Nonachlor	3.6	10	<3.6	<3.6	<3.6	
o,p'-DDT	6.1	18	<6.1	<6.1	<6.1	
p,p'-DDD	0.37	1.9	39	47	33	
Endosulfan-II	46	230	210	290	160	
p,p'-DDT	34	68	<34	<34	<34	
Endosulfan Sulfate	120	320	<120	<120	<120	
p,p'-Methoxychlor	9.4	47	11	<9.4	<9.4	
Mirex	0.52	2.6	< 0.5	< 0.5	< 0.5	
cis-Permethrin	62	180	<62	<62	<62	
trans-Permethrin	1.1	5.5	<1.2	<1.2	<1.2	
Total PCBs	110	310	170	200	310	
PBDE-28	6.0	10	< 6.0	< 6.0	11	
PBDE-47	21	49	28	28	23	
PBDE-99	13	27	<13	<13	<13	
PBDE-100	1.7	4.1	<1.7	<1.7	<1.7	
PBDE-153	1.3	2.2	<1.3	<1.3	<1.3	

386 MDL - method detection limit; MQL - method quantitation limit

387 Less than (<) values are results below the method detection limit.

Results in *italic* type are estimated values greater than the method detection limit but less than the method 388

389 quantitation limit. These values have a greater amount of uncertainty in the absolute value.

390 Results in **bold** type are reportable values greater than the method quantitation limit. 391 Table S6. Water concentrations of chlorinated pesticides, polychlorinated biphenyls (PCBs) and

392	polybrominated diphenyl ethers	s (PBDEs) estimated from	residues in semipermeable membrane
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devices buried in the sediment in Las Vegas Bay, Lake Mead, NV.	Vegas Bay, Lake Mead, NV.
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			Sampl	er Depth in S	ediment
	MDL	MQL	0-10 cm	10-20 cm	20-30 cm
	pg/L	pg/L	pg/L	pg/L	pg/L
Trifluralin	55	65	59	58	<55
Hexachlorobenzene (HCB)	3.5	18	6.2	5.6	6.9
Pentachloroanisole (PCA)	18	49	<18	<18	<18
Tefluthrin	200	240	210	<200	<200
alpha-Benzenehexachloride (a-BHC)	470	490	<470	<470	<470
Diazinon	97000	190000	<97000	<97000	<97000
Lindane	840	1100	<840	<840	<840
beta-Benzenehexachloride (b-BHC)	160	210	180	220	170
Heptachlor	11	20	<11	<11	<11
delta-Benzenehexachloride (d-BHC)	280	830	<280	<280	<280
Dacthal	70	180	<70	<70	<70
Chlorpyrifos	41	91	<41	<41	<41
Oxychlordane	3.6	18	<3.6	<3.6	<3.6
Heptachlor Epoxide	76	230	<76	<76	<76
trans-Chlordane	41	110	<41	<41	<41
trans-Nonachlor	94	260	<94	<94	<94
o,p'-DDE	54	150	<54	<54	71
cis-Chlordane	28	84	<28	<28	<28
Endosulfan	140	720	<140	<140	<140
p,p'-DDE	77	140	88	160	250
Dieldrin	68	140	<68	<68	<68
o,p'-DDD	31	83	<31	<31	<31
Endrin	54	91	57	<54	<54
cis-Nonachlor	3.9	19	6.3	<3.9	<3.9
o,p'-DDT	15	43	<15	<15	<15
p,p'-DDD	3.5	18	21	29	40
Endosulfan-II	300	1500	<300	<300	<300
p,p'-DDT	110	180	<110	<110	<110
Endosulfan Sulfate	500	1400	<500	<500	<500
p,p'-Methoxychlor	69	340	<69	<69	<69
Mirex	5.3	27	<5.3	<5.3	<5.3
cis-Permethrin	27	133	<27	<27	<27
trans-Permethrin	11	57	<11	<11	<11
Total PCBs	120	220	180	200	260
PBDE-28	23	360	<23	<23	<23
PBDE-47	110	270	<110	<110	<110
PBDE-99	6.1	12	<6.1	<6.1	<6.1
PBDE-100	33	54	<33	<33	<33
PBDE-153	26	64	<26	<26	38

394 MDL – method detection limit; MQL – method quantitation limit

Less than (<) values are results below the method detection limit.

396 Results in *italic* type are estimated values greater than the method detection limit but less than the method

397 quantitation limit. These values have a greater amount of uncertainty in the absolute value.

Results in **bold** type are reportable values greater than the method quantitation limit.

Table S7. Concentrations of waste indicator chemicals in polar organic chemical integrativesamplers (POCIS) deployed in the water column in Las Vegas Bay, Lake Mead, NV.

	Sampling			<u> </u>	Depth in Wate	
	Rate (R_s)	MDL	MQL	3.0 m	4.7 m	6.7 m
	L/d	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS
Tetrachloroethylene	NA	2.0	10	<2.0	<2.0	<2.0
Bromoform	NA	2.0	10	<2.0	<2.0	<2.0
Isopropylbenzene (cumene)	NA	2.0	10	<2.0	<2.0	<2.0
Phenol	0.097^{a}	10	50	<10	<10	110
1,4-Dichlorobenzene	NA	2.0	10	<2.0	<2.0	<2.0
d-Limonene	NA	2.0	10	<2.0	<2.0	<2.0
Acetophenone	0.082 ^a	2.0	10	<2.0	<2.0	<2.0
para-Cresol	0.087^{a}	20	100	<20	<20	13000
Isophorone	0.266 ^a	2.0	10	<2.0	<2.0	5.0
Camphor	NA	2.0	10	<2.0	<2.0	<2.0
Menthol	NA	10	50	<10	<10	<10
Naphthalene	NA	2.0	10	<2.0	<2.0	<2.0
Methyl salicylate	0.079 ^a	10	50	<10	<10	<10
Dichlorvos	NA	10	50	<10	<10	<10
Isoquinoline	NA	10	50	<10	<10	<10
Indole	0.085 ^a	10	50	<10	<10	680
2-Methyl naphthalene	NA	2.0	10	<2.0	<2.0	<2.0
1-Methyl naphthalene	NA	2.0	10	<2.0	<2.0	<2.0
2,6-Dimethylnaphthalene	NA	2.0	10	<2.0	<2.0	<2.0
Cashmeran (DPMI)	NA	2.0	10	<2.0	<2.0	<2.0
N,N-diethyltoluamide (DEET)	0.192 ^{a, b}	10	50	290	160	<10
Diethyl phthalate	0.061 ^a	2.0	10	110	100	55
p-tert-Octylphenol	0.058 ^c	10	50	95	65	<10
Benzophenone	0.067^{a}	2.0	10	10	<2.0	<2.0
Tributyl phosphate	0.150 ^a	2.0	10	65	40	<2.0
Ethyl citrate	0.265 ^a	10	50	50	35	<10
Cotinine	NA	10	50	<10	<10	<10
Celestolide (ADBI)	NA	2.0	10	<2.0	<2.0	<2.0
Prometon	NA	10	50	<10	<10	<10
Atrazine	NA	20	100	<20	<20	<20
Phantolide (AHMI)	NA	2.0	10	<2.0	<2.0	<2.0
4-Octylphenol	NA	20	100	<20	<20	<20
Tri(2-chloroethyl) phosphate	0.309 ^a	10	50	640	350	<10
N-butyl benzenesulfonamide	0.065 ^a	20	100	3100	2000	<20
Phenanthrene	NA	2.0	10	<2.0	<2.0	<2.0
Anthracene	NA	2.0	10	<2.0	<2.0	<2.0
Diazinon	NA	2.0	10	<2.0	<2.0	<2.0
Musk Ambrette	NA	2.0	10	<2.0	<2.0	<2.0
Carbazole	NA	2.0	10	<2.0	<2.0	<2.0
Caffeine	NA	10	50	<10	<10	<10
Traseolide (ATII)	NA	2.0	10	<2.0	<2.0	<2.0
Galaxolide (HHCB)	0.216 ^a	2.0	10	300	190	55
Tonalide (AHTN)	0.222 ^a	2.0	10	35	20	10
Musk Xylene	NA	2.0	10	<2.0	<2.0	<2.0

Carbaryl	NA	10	50	<10	<10	<10
Metalaxyl	NA	10	50	<10	<10	<10
Bromacil	NA	20	100	<20	<20	<20
Anthraquinone	NA	10	50	<10	<10	<10
Musk Ketone	NA	2.0	10	<2.0	<2.0	<2.0
Chlorpyrifos	NA	10	50	<10	<10	<10
Fluoranthene	NA	2.0	10	<2.0	<2.0	<2.0
Pyrene	NA	2.0	10	<2.0	<2.0	<2.0
Tri(dichloroisopropyl) phosphate	0.050^{a}	10	50	550	270	25
Tri(butoxyethyl) phosphate	NA	20	100	<20	<20	<20
Triphenyl phosphate	NA	2.0	10	<2.0	<2.0	<2.0
Diethylhexylphthalate (DEHP)	0.041^{a}	230	480	<230	<230	390
Benzo[a]pyrene	NA	2.0	10	<2.0	<2.0	<2.0
Cholesterol	NA	1000	2900	<1000	<1000	<1000

401 MDL – method detection limit; MQL – method quantitation limit

402 Less than (<) values are results below the method detection limit.

403 Results in *italic* type are estimated values greater than the method detection limit but less than the method

404 quantitation limit. These values have a greater amount of uncertainty in the absolute value.

405 Results in **bold** type are reportable values greater than the method quantitation limit.

406 NA – not applicable, sampling rate not needed as there were no detection for this chemical

407 ^a Alvarez, Unpublished Data

408 ^b Bartlet-Hunt et al., 2011

409 ^c Harman et al., 2008

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411

413 Table S8. Concentrations of waste indicator chemicals in polar organic chemical integrative

samplers (POCIS) buried in the sediment in Las Vegas Bay, Lake Mead, NV.

	Sampling			Sampler Depth in Sediment		
	Rate (R_s)	MDL	MQL	0-10 cm	10-20 cm	20-30 cm
	L/d	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS
Tetrachloroethylene	NA	2.0	10	<2.0	<2.0	<2.0
Bromoform	NA	2.0	10	<2.0	<2.0	<2.0
Isopropylbenzene (cumene)	NA	2.0	10	<2.0	<2.0	<2.0
Phenol	0.097 ^a	10	50	1300	640	<10
1,4-Dichlorobenzene	NA	2.0	10	<2.0	<2.0	<2.0
d-Limonene	NA	2.0	10	<2.0	<2.0	<2.0
Acetophenone	0.082^{a}	2.0	10	30	60	<2.0
para-Cresol	0.087^{a}	20	100	11000	6900	<20
Isophorone	0.266 ^a	2.0	10	40	70	<2.0
Camphor	NA	2.0	10	<2.0	<2.0	<2.0
Menthol	NA	10	50	<10	<10	<10
Naphthalene	NA	2.0	10	<2.0	<2.0	<2.0
Methyl salicylate	0.079 ^a	10	50	20	40	<10
Dichlorvos	NA	10	50	<10	<10	<10
Isoquinoline	NA	10	50	<10	<10	<10
Indole	0.085 ^a	10	50	950	760	<10
2-Methyl naphthalene	NA	2.0	10	<2.0	<2.0	<2.0
1-Methyl naphthalene	NA	2.0	10	<2.0	<2.0	<2.0
2,6-Dimethylnaphthalene	NA	2.0	10	<2.0	<2.0	<2.0
Cashmeran (DPMI)	NA	2.0	10	<2.0	<2.0	<2.0
N,N-diethyltoluamide (DEET)	0.192 ^{a, b}	10	50	200	<10	<10
Diethyl phthalate	0.061 ^a	2.0	10	490	240	<2.0
p-tert-Octylphenol	0.058 ^c	10	50	<10	<10	<10
Benzophenone	0.067 ^a	2.0	10	<2.0	<2.0	<2.0
Tributyl phosphate	0.150 ^a	2.0	10	<2.0	20	<2.0
Ethyl citrate	0.265 ^a	10	50	<10	<10	<10
Cotinine	NA	10	50	<10	<10	<10
Celestolide (ADBI)	NA	2.0	10	<2.0	<2.0	<2.0
Prometon	NA	10	50	<10	<10	<10
Atrazine	NA	20	100	<20	<20	<20
Phantolide (AHMI)	NA	2.0	10	<2.0	<2.0	<2.0
4-Octylphenol	NA	20	100	<20	<20	<20
Tri(2-chloroethyl) phosphate	0.309 ^a	10	50	<10	<10	<10
N-butyl benzenesulfonamide	0.065 ^a	20	100	<20	<20	<20
Phenanthrene	NA	2.0	10	<2.0	<2.0	<2.0
Anthracene	NA	2.0	10	<2.0	<2.0	<2.0
Diazinon	NA	2.0	10	<2.0	<2.0	<2.0
Musk Ambrette	NA	2.0	10	<2.0	<2.0	<2.0
Carbazole	NA	2.0	10	<2.0	<2.0	<2.0
Caffeine	NA	10	50	<10	<10	<10
Traseolide (ATII)	NA	2.0	10	<2.0	<2.0	<2.0
Galaxolide (HHCB)	0.216 ^a	2.0	10	130	<2.0 170	<2.0
Tonalide (AHTN)	0.222 ^a	2.0	10	30	20	<2.0
Musk Xylene	NA	2.0	10	<2.0	<2.0	<2.0

Carbaryl	NA	10	50	<10	<10	<10
Metalaxyl	NA	10	50	<10	<10	<10
Bromacil	NA	20	100	<20	<20	<20
Anthraquinone	NA	10	50	<10	<10	<10
Musk Ketone	NA	2.0	10	<2.0	<2.0	<2.0
Chlorpyrifos	NA	10	50	<10	<10	<10
Fluoranthene	NA	2.0	10	<2.0	<2.0	<2.0
Pyrene	NA	2.0	10	<2.0	<2.0	<2.0
Tri(dichloroisopropyl) phosphate	0.050^{a}	10	50	50	<10	<10
Tri(butoxyethyl) phosphate	NA	20	100	<20	<20	<20
Triphenyl phosphate	NA	2.0	10	<2.0	<2.0	<2.0
Diethylhexylphthalate (DEHP)	0.041 ^a	1000	2400	<1000	<1000	<1000
Benzo[a]pyrene	NA	2.0	10	<2.0	<2.0	<2.0
Cholesterol	NA	1200	2200	<1200	<1200	<1200

415 MDL – method detection limit; MQL – method quantitation limit

416 Less than (<) values are results below the method detection limit.

417 Results in *italic* type are estimated values greater than the method detection limit but less than the method

418 quantitation limit. These values have a greater amount of uncertainty in the absolute value.

419 Results in **bold** type are reportable values greater than the method quantitation limit.

420 NA – not applicable, sampling rate not needed as there were no detection for this chemical

421 ^a Alvarez, Unpublished Data

422 ^b Bartlet-Hunt et al., 2011

423 ^c Harman et al., 2008

424

- 426 Table S9. Nominal concentrations of pharmaceuticals and illicit drugs in polar organic chemical
- 427 integrative samplers (POCIS) deployed in the water column and buried in the sediment in Las
- 428 Vegas Bay, Lake Mead, NV.

	Sampling	Sampler I	Depth in Wate	er Column	Sampler Depth in Sediment		
	Rate (R_s)	3.0 m	4.7 m	6.7 m	0-10 cm	10-20 cm	20-30 cm
	L/d	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS
Azithromycin	NA	ND	ND	ND	ND	ND	ND
Clarithromycin	0.668 ^a	trace	ND	47	ND	ND	ND
Clindamycin	NA	ND	ND	ND	ND	ND	ND
Hydrocodone	0.050 ^b	170	ND	ND	ND	ND	ND
MDMA (Ecstasy)	NA	ND	ND	ND	ND	ND	ND
Methamphetamine	0.089 ^b	32	ND	ND	ND	ND	ND
N,N-DMPEA	NA	ND	ND	ND	ND	ND	ND
Pseudoephredrine	NA	ND	ND	ND	ND	ND	ND
Roxithromycin	NA	ND	ND	ND	ND	ND	ND
Urobilin	NA	ND	ND	ND	ND	ND	ND

429 ND – not detected.

430 Trace – identity of the clindamycin was confirmed by LC/MS/MS but the concentration was too low for

431 quantitation.

432 Results in **bold** type are reportable values greater than the method quantitation limit.

433 NA – not applicable, sampling rate not needed as there were no detection for this chemical

434 ^a MacLeod et al., 2007

435 ^b Alvarez, Unpublished Data

- 437 Table S10. Concentrations of performance reference compounds (PRCs) recovered from
- 438 semipermeable membrane devices (SPMDs) deployed in the water column and buried in the
- 439 sediment in Las Vegas Bay, Lake Mead, NV.

		Acenaphthylene- d_{10}	Acenaphthene- d_{10}	Fluorene- d_{10}	Phenanthrene- d_{10}	Pyrene- d_{10}	Dibenz[a,h]anthracene- d_{14}^{a}
		ng/SPMD	ng/SPMD	ng/SPMD	ng/SPMD	ng/SPMD	ng/SPMD
Water Column	Fabrication Blank	730	790	880	1030	1250	1060
	Field Blank	680	750	887	1070	1240	1050
	Blank Average ^b	705	770	884	1050	1245	1055
	3.0 m	0	0	0	60	550	930
Wa	4.7 m	0	0	0	40	490	950
	6.7 m	160	350	400	630	1070	1160
г	Fabrication Blank	270	300	340	430	590	670
eptl	Field Blank	270	320	370	430	460	560
Sediment Depth	Blank Average ^b	270	310	355	430	525	615
	0-10 cm	150	330	370	460	600	650
	10-20 cm	100	210	260	400	550	630
	20-30 cm	90	210	250	360	540	600

440 ^a Dibenz[a,h]anthracene- d_{14} was used as a photolysis marker.

441 ^b The average of the PRC values from the blanks was used to determine the initial PRC concentrations (N_0) .

Table S11. Recovery of priority pollutant polycyclic aromatic hydrocarbons (PAHs) from

semipermeable membrane devices (SPMDs) processed concurrently with field SPMDs deployed

in the water column and buried in the sediment in Las Vegas Bay, Lake Mead, NV.

	Water Column	Sediment Depth
	% Recovery	% Recovery
Naphthalene	41	28
Acenaphthylene	67	51
Acenaphthene	70	55
Fluorene	79	63
Phenanthrene	87	73
Anthracene	85	73
Fluoranthene	92	84
Pyrene	93	84
Benz[a]anthracene	94	91
Chrysene	94	90
Benzo[b]fluoranthene	94	89
Benzo[k]fluoranthene	92	87
Benzo[a]pyrene	93	87
Indeno[1,2,3-c,d]pyrene	96	85
Dibenz[<i>a</i> , <i>h</i>]anthracene	94	79
Benzo[g,h,i]perylene	91	83

446

448 Table S12. Recovery of chlorinated pesticides, polychlorinated biphenyls (PCBs) and

449 polybrominated diphenyl ethers (PBDEs) from semipermeable membrane devices (SPMDs)

450 processed concurrently with field SPMDs deployed in the water column and buried in the

451 sediment in Las Vegas Bay, Lake Mead, NV.

% Recovery % Recovery Trifluralin 12 13 Hexachlorobenzene (HCB) 91 70 Pentachloroanisole (PCA) 96 70 Tefluthrin 14 11 alpha-Benzenehexachloride (a-BHC) 74 70 Diazinon 22 8.9 Lindane 82 80 beta-Benzenehexachloride (b-BHC) 79 77 Heptachlor 87 86 delta-Benzenehexachloride (d-BHC) 74 74 Dacthal 29 26 Chloryprifos 45 22 Oxychlordane 98 96 trans-Chlordane 88 86 trans-Chlordane 87 94 Endosulfan 85 99 p.p'-DDE 83 87 Dieldrin 86 88 o.p'DDT 79 85 p.p'-DDD 94 84 Endosulfan-II 83 49 p.p'-DDT <t< th=""><th></th><th>Water Column</th><th>Sediment Depth</th></t<>		Water Column	Sediment Depth																																																																																																
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PCBs9189</td><td>Dieldrin</td><td>86</td><td>88</td></tr> <tr><td>cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>o,p'-DDD</td><td>96</td><td>90</td></tr> <tr><td>o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>Endrin</td><td>92</td><td>94</td></tr> <tr><td>p,p'-DDD 94 84 Endosulfan-II 83 49 p,p'-DDT 83 79 Endosulfan Sulfate 79 49 p,p'-Methoxychlor 30 44 Mirex 88 83 cis-Permethrin 4.5 5.7 trans-Permethrin 2.0 3.4 Total PCBs 91 89</td><td>cis-Nonachlor</td><td>77</td><td>71</td></tr> <tr><td>Endosulfan-II 83 49 p,p'-DDT 83 79 Endosulfan Sulfate 79 49 p,p'-Methoxychlor 30 44 Mirex 88 83 cis-Permethrin 4.5 5.7 trans-Permethrin 2.0 3.4 Total PCBs 91 89</td><td>o,p'-DDT</td><td>79</td><td>85</td></tr> <tr><td>p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>p,p'-DDD</td><td>94</td><td>84</td></tr> <tr><td>Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>Endosulfan-II</td><td>83</td><td>49</td></tr> <tr><td>p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>p,p'-DDT</td><td>83</td><td>79</td></tr> <tr><td>Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>Endosulfan Sulfate</td><td>79</td><td>49</td></tr> <tr><td>cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189</td><td>p,p'-Methoxychlor</td><td>30</td><td>44</td></tr> <tr><td>trans-Permethrin2.03.4Total PCBs9189</td><td>Mirex</td><td>88</td><td>83</td></tr> <tr><td>Total PCBs9189</td><td>cis-Permethrin</td><td>4.5</td><td>5.7</td></tr> <tr><td></td><td>trans-Permethrin</td><td>2.0</td><td>3.4</td></tr> <tr><td>PBDE-28 88 93</td><td>Total PCBs</td><td>91</td><td>89</td></tr> <tr><td></td><td>PBDE-28</td><td>88</td><td>93</td></tr>	Dacthal	29	26	Heptachlor Epoxide 98 96 trans-Chlordane 88 86 trans-Nonachlor 86 84 o,p'-DDE 97 108 cis-Chlordane 87 94 Endosulfan 85 99 p,p'-DDE 83 87 Dieldrin 86 88 o,p'-DDD 96 90 Endrin 92 94 cis-Nonachlor 77 71 o,p'-DDD 96 90 Endrin 92 94 cis-Nonachlor 77 71 o,p'-DDT 79 85 p,p'-DDT 83 49 p,p'-DDT 83 49 p,p'-DDT 83 79 Endosulfan-II 83 49 p,p'-DDT 83 44 Mirex 88 83 cis-Permethrin 4.5 5.7 trans-Permethrin 2.0 3.4 Total PCBs 91 89	Chlorpyrifos	45	22	trans-Chlordane 88 86 trans-Nonachlor 86 84 o,p'-DDE 97 108 cis-Chlordane 87 94 Endosulfan 85 99 p,p'-DDE 83 87 Dieldrin 86 88 o,p'-DDD 96 90 Endrin 92 94 cis-Nonachlor 77 71 o,p'-DDT 79 85 p,p'-DDT 83 49 p,p'-DDD 94 84 Endosulfan-II 83 49 p,p'-DDT 83 79 Endosulfan-Sulfate 79 49 p,p'-DDT 83 79 Endosulfan Sulfate 79 49 p,p'-Methoxychlor 30 44 Mirex 88 83 cis-Permethrin 4.5 5.7 trans-Permethrin 2.0 3.4 Total PCBs 91 89	Oxychlordane	98	91	trans-Nonachlor8684o,p'-DDE97108cis-Chlordane8794Endosulfan8599p,p'-DDE8387Dieldrin8688o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDT8349p,p'-DDT8379Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	Heptachlor Epoxide	98	96	o,p'-DDE97108cis-Chlordane8794Endosulfan8599p,p'-DDE8387Dieldrin8688o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin2.03.4Total PCBs9189	trans-Chlordane	88	86	cis-Chlordane8794Endosulfan8599p,p'-DDE8387Dieldrin8688o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	trans-Nonachlor	86	84	Endosulfan8599p,p'-DDE8387Dieldrin8688o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	o,p'-DDE	97	108	p,p'-DDE8387Dieldrin8688o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	cis-Chlordane	87	94	Na8688o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	Endosulfan	85	99	o,p'-DDD9690Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	p,p'-DDE	83	87	Endrin9294cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	Dieldrin	86	88	cis-Nonachlor7771o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	o,p'-DDD	96	90	o,p'-DDT7985p,p'-DDD9484Endosulfan-II8349p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	Endrin	92	94	p,p'-DDD 94 84 Endosulfan-II 83 49 p,p'-DDT 83 79 Endosulfan Sulfate 79 49 p,p'-Methoxychlor 30 44 Mirex 88 83 cis-Permethrin 4.5 5.7 trans-Permethrin 2.0 3.4 Total PCBs 91 89	cis-Nonachlor	77	71	Endosulfan-II 83 49 p,p'-DDT 83 79 Endosulfan Sulfate 79 49 p,p'-Methoxychlor 30 44 Mirex 88 83 cis-Permethrin 4.5 5.7 trans-Permethrin 2.0 3.4 Total PCBs 91 89	o,p'-DDT	79	85	p,p'-DDT8379Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	p,p'-DDD	94	84	Endosulfan Sulfate7949p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	Endosulfan-II	83	49	p,p'-Methoxychlor3044Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	p,p'-DDT	83	79	Mirex8883cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	Endosulfan Sulfate	79	49	cis-Permethrin4.55.7trans-Permethrin2.03.4Total PCBs9189	p,p'-Methoxychlor	30	44	trans-Permethrin2.03.4Total PCBs9189	Mirex	88	83	Total PCBs9189	cis-Permethrin	4.5	5.7		trans-Permethrin	2.0	3.4	PBDE-28 88 93	Total PCBs	91	89		PBDE-28	88	93
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trans-Permethrin2.03.4Total PCBs9189	Mirex	88	83																																																																																																
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	trans-Permethrin	2.0	3.4																																																																																																
PBDE-28 88 93	Total PCBs	91	89																																																																																																
	PBDE-28	88	93																																																																																																

PBI	DE-47	94	117
PBI	DE-99	44	57
PBI	DE-100	39	49
PBI	DE-153	3.3	11

454 Table S13. Concentrations of polycyclic aromatic hydrocarbons (PAHs) from semipermeable

membrane device (SPMD) blanks processed concurrently with field SPMDs deployed in the
water column and buried in the sediment in Las Vegas Bay, Lake Mead, NV.

	Water Column Fabrication Field			Sediment Depth Fabrication Field			
	Lab Blank	Blank	Blank	Lab Blank	Blank	Blank	
	ng/SPMD	ng/SPMD	ng/SPMD	ng/SPMD	ng/SPMD	ng/SPMD	
Naphthalene	0	0	0	10	0	10	
Acenaphthylene	0	0	0	0	0	0	
Acenaphthene	0	0	0	0	0	0	
Fluorene	0	0	0	0	0	0	
Phenanthrene	20	10	10	10	10	10	
Anthracene	0	0	0	0	0	0	
Fluoranthene	10	0	0	10	0	0	
Pyrene	0	0	0	0	0	0	
Benz[a]anthracene	0	0	0	0	0	0	
Chrysene	0	0	0	0	0	0	
Benzo[b]fluoranthene	0	0	0	0	0	0	
Benzo[k]fluoranthene	0	0	0	0	0	0	
Benzo[<i>a</i>]pyrene	0	0	0	0	0	0	
Indeno[1,2,3- <i>c</i> , <i>d</i>]pyrene	0	0	0	0	0	0	
Dibenz[<i>a</i> , <i>h</i>]anthracene	0	0	0	0	0	0	
Benzo[g,h,i]perylene	0	0	0	0	0	0	
Benzo[b]thiophene	0	0	0	0	0	0	
2-methylnaphthalene	10	10	0	10	0	0	
1-methylnaphthalene	0	0	0	0	0	0	
Biphenyl	0	0	0	0	0	0	
1-ethylnaphthalene	0	0	0	0	0	0	
1,2-dimethylnaphthalene	0	0	0	0	0	0	
4-methylbiphenyl	0	0	0	0	0	0	
2,3,5-trimethylnaphthalene	0	0	0	0	0	0	
1-methylfluorene	20	10	20	10	0	0	
Dibenzothiophene	0	0	0	0	0	0	
2-methylphenanthrene	0	0	0	0	0	0	
9-methylanthracene	0	0	0	0	0	0	
3,6-dimethylphenanthrene	0	0	0	0	0	0	
2-methylfluoranthene	0	0	0	0	0	0	
Benzo[b]naphtho[2,1-d]thiophene	0	0	0	0	0	0	
Benzo[e]pyrene	0	0	0	0	0	0	
Perylene	0	0	0	0	0	0	
3-methylcholanthrene	0	0	0	0	0	0	

- 457 Table S14. Concentrations of chlorinated pesticides, polychlorinated biphenyls (PCBs) and
- polybrominated diphenyl ethers (PBDEs) from semipermeable membrane device (SPMD) blanks
 processed concurrently with field SPMDs deployed in the water column and buried in the
- processed concurrently with field SPMDs deployed in the water column and buried in thesediment in Las Vegas Bay, Lake Mead, NV.

	V Lab Blank ng/SPMD	Vater Column Fabrication Blank ng/SPMD	Field Blank ng/SPMD	S Lab Blank ng/SPMD	ediment Depth Fabrication Blank ng/SPMD	Field Blank ng/SPMD
Trifluralin	4.5	4.9	4.5	2.6	2.6	2.7
Hexachlorobenzene (HCB)	0.18	0.08	0.28	0.06	0.03	0.05
Pentachloroanisole (PCA)	5.1	1.6	0.54	0.36	0.47	0.00
Tefluthrin alpha-Benzenehexachloride	12	10	5.0	5.6	5.9	5.8
(a-BHC)	5.8	5.7	5.2	3.0	3.0	3.0
Diazinon	36	35	33	17	22	27
Lindane beta-Benzenehexachloride	6.7	6.7	6.9	3.2	3.3	3.5
(b-BHC)	1.9	1.9	1.4	0.99	0.91	0.94
Heptachlor delta-Benzenehexachloride	0.88	1.0	2.6	0.51	0.53	0.53
(d-BHC)	0	0	3.3	1.6	0	0
Dacthal	0.78	0.74	0	0.14	0.26	0.64
Chlorpyrifos	1.1	1.1	0	0.24	0.20	0.40
Oxychlordane	0	0	0	0	0	0
Heptachlor Epoxide	2.0	2.1	0	0	0.93	0
trans-Chlordane	0.32	0.3	0.86	0.17	0.16	1.2
trans-Nonachlor	0.74	0.9	1.6	0.54	0.50	2.5
o,p'-DDE	0.78	0.8	2.7	0.40	0.42	1.7
cis-Chlordane	0	0	0.10	0	0	0.74
Endosulfan	0	0	0	0	0	0
p,p'-DDE	4.7	6.2	6.2	2.3	2.7	3.2
Dieldrin	1.4	0	1.9	0.71	1.0	1.3
o,p'-DDD	0	3.1	1.9	0	0.56	0.78
Endrin	0	1.1	0	1.1	1.3	1.0
cis-Nonachlor	0	0.94	0.40	0.05	0.02	0
o,p'-DDT	0	0	1.6	0	0	0.40
p,p'-DDD	0	0	0	0	0	0
Endosulfan-II	0	0	0	0	0	0
p,p'-DDT	8.4	8.6	13	4.0	4.0	5.0
Endosulfan Sulfate	0.08	0.04	0.38	0	0.12	0.24
p,p'-Methoxychlor	0	0	0	0	0	0
Mirex	0	0	0	0	0	0
cis-Permethrin	2.1	0	85	0	0	0

				I		
trans-Permethrin	0	0	0	0	0	0
Total PCBs	7.0	4.5	29	3.6	2.9	4.3
PBDE-28	2.1	2.2	2.6	0.96	0.92	1.1
PBDE-47	2.8	7.0	4.9	3.6	1.6	1.3
PBDE-99	2.0	3.6	3.0	1.1	0.81	0.90
PBDE-100	0.24	0.44	0.18	0.18	0.12	0.13
PBDE-153	0.22	0.28	0.28	0.17	0.44	0.21

- 463 Table S15. Concentrations of waste indicator chemicals in polar organic chemical integrative
- samplers (POCIS) blanks processed concurrently with field SPMDs deployed in the water

	Lab Blank	Water Column Fabrication Blank	Field Blank	Se Lab Blank	ediment Depth Fabrication Blank	Field Blank
	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS	ng/POCIS
Tetrachloroethylene	0	0	0	0	0	0
Bromoform	0	0	0	0	0	0
Isopropylbenzene (cumene)	0	0	0	0	0	0
Phenol	0	0	0	0	0	0
1,4-Dichlorobenzene	0	0	0	0	0	0
d-Limonene	0	0	0	0	0	0
Acetophenone	0	0	0	0	0	0
para-Cresol	0	0	0	0	0	0
Isophorone	0	0	0	0	0	0
Camphor	0	0	0	0	0	0
Menthol	0	0	0	0	0	0
Naphthalene	0	0	0	0	0	0
Methyl salicylate	0	0	0	0	0	0
Dichlorvos	0	0	0	0	0	0
Isoquinoline	0	0	0	0	0	0
Indole	0	0	0	0	0	0
2-Methyl naphthalene	0	0	0	0	0	0
1-Methyl naphthalene	0	0	0	0	0	0
2,6-Dimethylnaphthalene	0	0	0	0	0	0
Cashmeran (DPMI)	0	0	0	0	0	0
N,N-diethyltoluamide (DEET)	0	0	0	0	0	0
Diethyl phthalate	0	0	0	0	0	0
p-tert-Octylphenol	0	0	0	0	0	0
Benzophenone	0	0	0	0	0	0
Tributyl phosphate	0	0	0	0	0	0
Ethyl citrate	0	0	0	0	0	0
Cotinine	0	0	0	0	0	0
Celestolide (ADBI)	0	0	0	0	0	0
Prometon	0	0	0	0	0	0
Atrazine	0	0	0	0	0	0
Phantolide (AHMI)	0	0	0	0	0	0
4-Octylphenol	0	0	0	0	0	0
Tri(2-chloroethyl) phosphate	0	0	0	0	0	0
N-butyl benzenesulfonamide	0	0	0	0	0	0

465 column and buried in the sediment in Las Vegas Bay, Lake Mead, NV.

Phenanthrene	0	0	0	0	0	0
Anthracene	0	0	0	0	0	0
Diazinon	0	0	0	0	0	0
Musk Ambrette	0	0	0	0	0	0
Carbazole	0	0	0	0	0	0
Caffeine	0	0	0	0	0	0
Traseolide (ATII)	0	0	0	0	0	0
Galaxolide (HHCB)	0	0	0	0	0	0
Tonalide (AHTN)	0	0	0	0	0	0
Musk Xylene	0	0	0	0	0	0
Carbaryl	0	0	0	0	0	0
Metalaxyl	0	0	0	0	0	0
Bromacil	0	0	0	0	0	0
Anthraquinone	0	0	0	0	0	0
Musk Ketone	0	0	0	0	0	0
Chlorpyrifos	0	0	0	0	0	0
Fluoranthene	0	0	0	0	0	0
Pyrene Tri(dichloroisopropyl)	0	0	0	0	0	0
phosphate	0	0	0	0	0	0
Tri(butoxyethyl) phosphate	0	0	0	0	0	0
Triphenyl phosphate	0	0	0	0	0	0
Diethylhexylphthalate (DEHP)	150	130	80	230	610	480
Benzo[a]pyrene	0	0	0	0	0	0
Cholesterol	520	0	190	580	850	790

