



## Estimation of atmospheric PCB releases from industrial facilities in Turkey

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### ABSTRACT

PCB production reduced or stopped in most countries in the late 1970s. According to the Stockholm Convention all the party countries have to list their PCB containing equipment and prepare national inventories. A national implementation plan was prepared in Turkey however, this plan is inadequate. Furthermore, there has not been any information prepared about stationary emission sources, contributing to atmospheric PCB inputs so far. It is aimed in this study to meet this deficit. This study will also help to make stronger comments on identifying sources of atmospheric PCB concentrations. PCB emission factors were utilized to determine the emissions. An average of 920 kg yr<sup>-1</sup> of PCB emission is estimated in this study within the borders of Turkey. Highest contribution occurs in Kocaeli located on the North–West of the country by 47% of the overall emissions. Secondary most pollutant site is determined to be Aliaga on the west of the country. Both of this regions are heavily, industrialized and several different industrial processes are present. Iron–steel manufacturing and copper smelting processes comprises the 94% of the PCB releases from stationary sources. A simple box model was applied at two hot spots to determine potential contribution of the estimated emissions to atmospheric concentrations. In Dilovasi, the average contribution from industrial sources was determined as 244 pg m<sup>-3</sup> while the average contribution for Aliaga was calculated as 126 pg m<sup>-3</sup> within a 50 km x 50 km grid.

**Keywords:** PCB emission inventory, emission factor, Krigging method, simple box model

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### 1. Introduction

Manufacture and commercial use of polychlorinated biphenyls (PCBs) started in 1929. Environmental contamination of these compounds was first detected by Jensen (1966). Due to the fact that these compounds are toxic and resistant to any kind of degradation processes, a great number of manufacturers reduced or halted PCB production during 1970s (Falandysz and Szymczyk, 2001), however, some countries continued until 1990s (Wyrzykowska et al., 2006). Breivik et al. (2002b) estimated that 1.3 million tones of PCBs have been produced so far and about 440 to 92 000 tons of PCBs have been emitted to the environment. Although the use of PCBs are not allowed in most countries today, emissions take place continuously from previously used equipments and waste sites (Breivik et al., 2002a).

Throughout the past years, nearly all PCB emissions took place between northern temperate latitudes which are between 30° and 60°N (Meijer et al., 2003). The use of PCBs between these latitudes has been estimated to be as approximately 86% of global usage and consequently these regions have the highest share of atmospheric emissions (Wyrzykowska et al., 2006). On the other hand first measures of diminishing their manufacture and use were firstly applied between those latitudes in Western Europe, Japan and America in the 1970s (Wyrzykowska et al., 2006). A newer study by Karaca et al. (2009) in Istanbul, Turkey revealed that the region is prone to long–range transport of pollutants from Mediterranean Region, Europe, and Asia. Located between 36° and

42° Northern latitudes, Turkey could be considered being under the effect of long–range transport of pollutants from these regions.

Because of their toxic and persistent nature and carcinogenic/mutagenic properties, PCBs are regarded as one of the twelve persistent organic pollutants (POPs) of the Stockholm Convention which was adopted in 2001 (UNEP, 2001). The Stockholm Convention brought some responsibilities to the countries which are party to the agreement. Responsibility of the countries includes establishment of inventories on the level of the POPs, identification of previously polluted sites and conduct environmental management of POP containing wastes. Gedik and Imamoglu (2010) expressed the fact that countries which are party to Stockholm Convention are required to identify pollution hot spots to execute environmental remediation of the polluted sites. Although Turkey has never been a PCB producing country, the use of PCBs was prohibited in Turkey in 1995. However, it is known that Turkey had already imported PCB–containing equipment in the past. According to National Implementation Plan prepared in 2004, 1 972 capacitors and 290 transformers are filled with PCB containing oil. As an estimation, approximately 5 000 tons of PCB load was thought to be present in Turkey (Ministry of Environment and Forestry, 2006). In the preparation of this inventory, a questionnaire was sent to the probable PCB holders to declare their PCB containing equipment. Forty seven percent of the transformers were investigated according to their label information, site inspections were done with test kits for only 100 transformers (Ministry of Environment and Forestry, 2006). Apart from Turkey, Hsu et al. (2003) stated that PCB emission

sources are not well-known and emissions, based on existing inventories, are drastically underestimated.

Ambient PCBs mostly originate from burning of PCB-containing materials, from air/water and air/soil exchange processes, from sludge handling processes, and solid waste handling operations (Tasdemir et al., 2005). Apart from these sources and intentional manufacturing, Erickson (2001) stated that PCB emissions also take place during production of chlorinated solvents, chlorophthalenes, chlorophenylsiloxane adhesives, organosilicone drugs, organic intermediates and pigments as well as during thermal processes such as waste combustion.

A number of research projects related to PCBs in various media have been conducted so far in Turkey, and Gedik and Imamoglu (2010) presented a comprehensive review through which they concluded that PCB concentrations were higher especially in heavily industrialized regions. Odabasi et al. (2009) reported ambient concentrations of PCBs for Aliaga, Turkey. They stated that highest concentrations were observed nearby an iron-steel manufacturing facility.

According to Weber et al. (2008), sustainable prevention of sites, which are dealing with especially POPs, from contamination, could be accomplished by integrated pollution prevention and control approach. As PCBs are not being produced anymore, unintentional releases, such as through industrial activities, must not be ignored. There is an important lack of PCB data measured at different media in Turkey (Salihoglu and Tasdemir, 2009). Gedik and Imamoglu (2010) pointed out that the results of PCB pollution in different regions of Turkey cannot be compared to national background levels due to the lack of previous monitoring studies in different environmental media. At least, continuous background pollution data has to be known to make a more precise evaluation of monitoring results. Therefore, thorough identification of atmospheric PCB sources is of critical importance for the implementation of total maximum daily load (TMDL) (Du and Rodenburg, 2007).

The objectives of this study are to (i) determine the continuous PCB releases from stationary sources with the goal of hot spot identification that will help the local authorities to locate pollution sources and take measures for sustainable prevention of the exposed sites, (ii) form the emission inventory of Turkey from production activities, (iii) visualize continuous background PCB emissions to help locating potential sources for further scientific ambient observation studies, and (iv) model hot spot zones to evaluate their potential impact.

## 2. Methodology

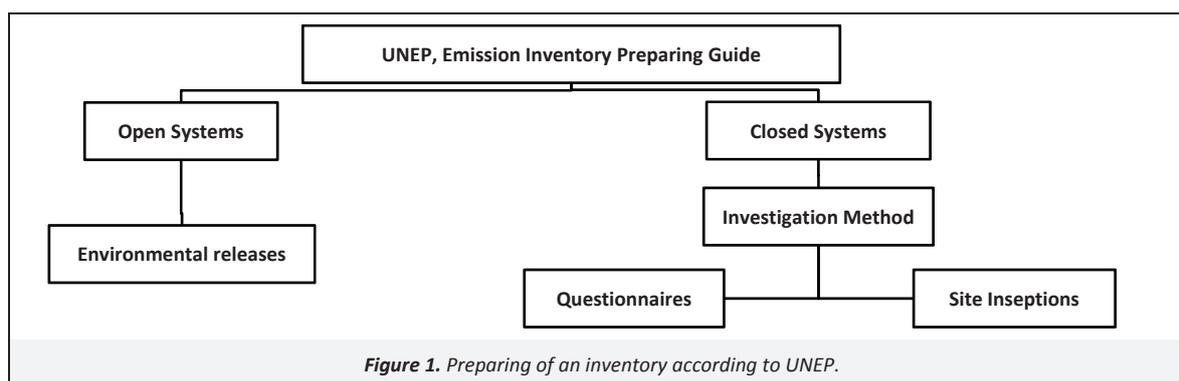
### 2.1. Inventory and emission factors

The steps involved in preparation scheme for an emission inventory from the Training Manual of Secretariat of the Basel Convention (UNEP, 2003) are shown in Figure 1.

The preparation scheme defines two distinct pathways for open and closed systems. Where, closed systems refer to equipments which were previously loaded with PCB containing materials such as, dielectric fluids, transformers capacitors and electromagnets. Open systems are the ones where the PCB containing material is not isolated by a controlled volume. Inks, lubricants, waxes and insulators are typical examples of this type. Also, in the Inventory Preparation Guidebook (UNEP, 2003) it is emphasized that as the book is related with closed systems, different investigation methods can be followed for open application involving environmental releases. For such an aim utilizing the emission factors is essential. Sullivan and Woods (2000) used emission factors from U.S. EPA (1995) for characterizing heavy metal emissions from sewage sludge incinerators. In another study, Ilek and Elbir (2012) estimated pollutant emission from transportation ferries by using emission factors of EMEP/CORINAIR Emission Inventory Guidebook (EEA, 2007) and modeled their contribution to ambient air quality within Izmir Bay. An inventory for closed systems should first be prepared in accordance with the National Implementation Plan. Drawbacks of this inventory were stated previously. However, for open systems or industrial applications any inventory has not been prepared yet. Environmental releases of PCBs are calculated according to emission factors available in the literature.

EPA's AP42 Compilation of Air Pollutant Emission Factors (U.S. EPA, 1995) and EEA (2007) were searched to identify PCB emitting industries. In Emission Inventory Guidebook of EMEP/CORINAIR (EEA, 2007) PCB emissions from cement manufacturing was listed as  $1 \mu\text{g ton}^{-1}$  cement produced. However, PCB emission factor for cement production is not contained in the latest Emission Inventory Guidebook (EEA, 2009). Instead, PCB emission factor of cement manufacturing is listed in "combustion in manufacturing industries and construction" section. POPs and as well as some other pollutants are originated through the combustion of fossil fuels. As being an integral part of the production system these pollutants are not discarded totally from production emission factors. The only thing done was listing the pollutant in the appropriate formation source. Therefore, emission factors of such pollutants are addressed in the combustion emission factors list. In the revised version, emission factor was stated as  $1 \mu\text{g ton}^{-1}$  clinker produced for cement production.

For the steel production plants, emission factors depend on the production activity. Each steel production technology, i.e., sinter production, electric arc furnaces and electric arc furnaces with pre-heaters have different emission factors. For the cement production and sinter production, values presented in EMEP/CORINAIR Emission Inventory Guidebook, 2009 was used. For electric arc furnaces and electric arc furnaces with pre-heaters, emission factors were taken from Odabasi et al. (2009). In that study, they generated emission factors from the stack gas measurements and steel production amounts of iron-steel plants in Aliaga, Turkey.



Other smelting process emission factors for pig iron, copper and zinc productions are available in EMEP/CORINAIR Emission Inventory Guidebook and these values were used in the present study.

Another emission factor is available for coal combustion. In the Emission Inventory Guidebook, emission factor for hard coal is stated under the section of “Combustion in Energy Industries”. Emission factor data is not available for brown coal; however under the section of “Combustion in Manufacturing Industries” for both brown and hard coal an emission factor is included. This value was utilized to calculate emissions generated from thermal power plants. All emission factors are listed in Table 1.

**Uncertainties.** The term of uncertainty refers lack of knowledge regarding the true value of a fixed but unknown amount. For inventory estimations it is recommended to state the uncertainty within the 95% confidence interval. If standard deviation of a set of experiments is less than 30%, then this distribution is regarded as normal distribution. For a normal distribution, 95% confidence

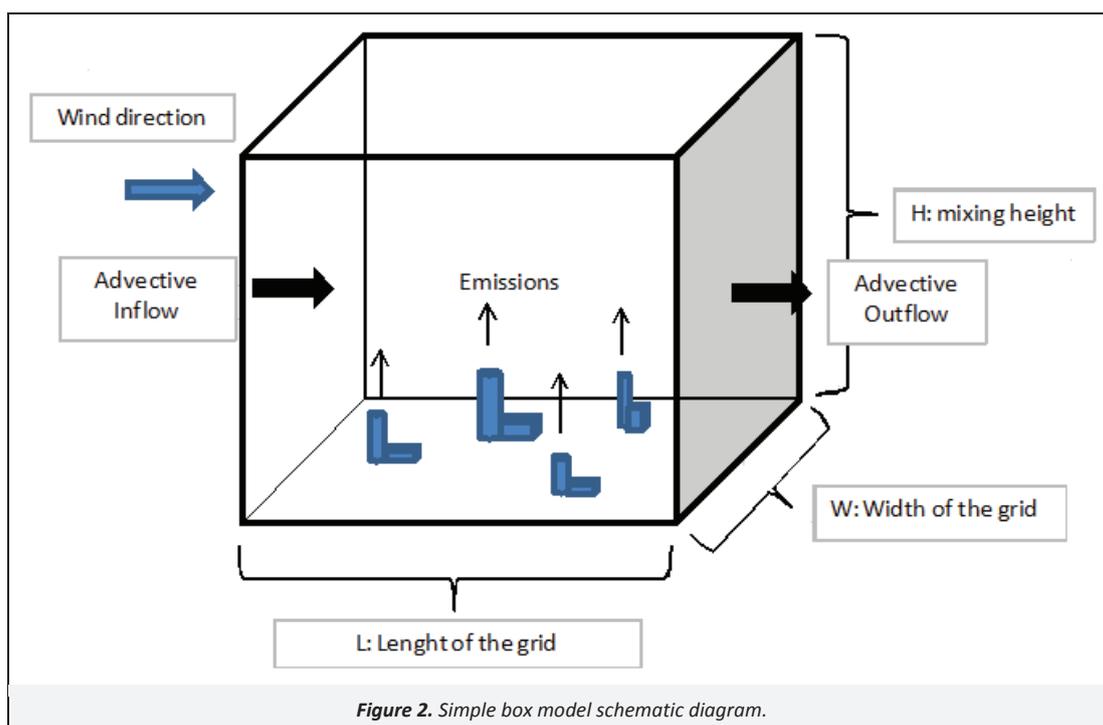
interval lies between  $\pm 2\sigma$  around the mean value (EEA, 2009). EMEP/CORINAIR emission factors include values of 95% confidence interval. For the factors which do not have any given value beside the mean value, error ranges specified in EMEP/CORINAIR–uncertainties section were selected. It was suggested to select a range from 50% to 100% of error for those which a number of measurements are small. Uncertainty from activity levels also have to be considered. However, a typical 0% of uncertainty for activity level is proposed for official statistics of activity data.

**2.2. Simple box model**

Simple box models are approximate since they include many assumptions. However they are an easy way of estimating atmospheric concentrations over an area source. A schematic diagram of a box model including source emissions, advective inflow and outflow according to the wind direction, dimensions of the box either horizontal and vertical (mixing height) axis is shown in Figure 2.

**Table 1.** Emission factors for diverse production types

Plant	Production type	Emission factor	95% confidence interval		Reference
			Lower	Upper	
Cement production		1 $\mu\text{g ton}^{-1}$	0.5 $\mu\text{g ton}^{-1}$	10 $\mu\text{g ton}^{-1}$	EEA (2009)
Steel production	Electric arc furnace	5.4 $\text{mg ton}^{-1}$	2.7 $\text{mg ton}^{-1}$	10.8 $\text{mg ton}^{-1}$	Odabasi et al. (2009)
Steel production	Electric arc furnace with pre-heating	171 $\text{mg ton}^{-1}$	85.5 $\text{mg ton}^{-1}$	342 $\text{mg ton}^{-1}$	Odabasi et al. (2009)
Steel production	Sinter production	3.6 $\text{mg ton}^{-1}$	1 $\text{mg ton}^{-1}$	13 $\text{mg ton}^{-1}$	EEA (2009)
Pig iron	Smelting	2 $\text{mg ton}^{-1}$	1.6 $\text{mg ton}^{-1}$	2.7 $\text{mg ton}^{-1}$	EEA (2009)
Copper production	Smelting	0.9 $\text{g ton}^{-1}$	0.6 $\text{g ton}^{-1}$	1.5 $\text{g ton}^{-1}$	EEA (2009)
Zinc production	Smelting	0.9 $\text{g ton}^{-1}$	0.3 $\text{g ton}^{-1}$	2.8 $\text{g ton}^{-1}$	EEA (2009)
Clinical Waste Incineration	Combustion	0.02 $\text{g ton}^{-1}$	0.002 $\text{g ton}^{-1}$	0.2 $\text{g ton}^{-1}$	EEA (2009)
Hard coal	Combustion	170 $\mu\text{g GJ}^{-1}$	85 $\mu\text{g GJ}^{-1}$	260 $\mu\text{g GJ}^{-1}$	EEA (2009)
Brown coal	Combustion	170 $\mu\text{g GJ}^{-1}$	85 $\mu\text{g GJ}^{-1}$	260 $\mu\text{g GJ}^{-1}$	EEA (2009)



**Figure 2.** Simple box model schematic diagram.

Assumptions made during the derivation of the model are (i) uniform mixing is present within the box whose horizontal boundaries enclose the urban area of interest, (ii) the model can predict only the volume-averaged concentration as a function of time, (iii) diffusion from individual sources is not considered, (iv) wind speed and direction is constant and its value does not change with the altitude, (v) back-ground concentration and emissions within the box are constant, and (vi) no formation or destruction of the chemical species occurs.

For simple box model without chemical transformations, one can derive the concentration by considering the mass conservation within the box. There are two major inputs to the box and one output from the box. Emissions from unit area and advection of the polluted air by the wind are the inputs to the box, whereas advection by the wind which is polluted from the emissions within the box and from the transported air from upwind direction is the output of the box. Derived simple box model formula considering the mass conservation is given in Equation (1):

$$C = b + \frac{q \cdot L}{u \cdot H} \tag{1}$$

where, the terms *b*, *q*, *L*, *u* and *H* correspond to back-ground concentration ( $\text{pg m}^{-3}$ ), emission rate per area ( $\text{pg m}^{-2} \text{s}^{-1}$ ), length (m), average wind speed ( $\text{m s}^{-1}$ ) and mixing height (m), respectively.

The required meteorological parameters; wind speed and wind direction was gathered from nearby meteorological stations of Turkish State Meteorological Service. The reported average wind speed value for Dilovasi and Aliaga are 2.75 and 3.00  $\text{m s}^{-1}$ , respectively. Wind directions for both sites are shown in Figure 3.

The mixing height value was taken from the ARL archived meteorological data of NOAA (NOAA, 2012). The mixing height values are 410 m and 480 m for Dilovasi and Aliaga, respectively.

### 3. Results and Discussion

#### 3.1. Emission estimates

Once grid map of 50 km x 50 km was generated, PCB releases within each cell were calculated in accordance with Equation (2):

$$Emission = Capacity \times Emission\ Factor \tag{2}$$

Activity levels used through the calculation process of the total emissions are provided in Table 2.

Production capacities of the facilities together with corresponding emission factors were utilized in the estimation of annual atmospheric PCB inputs. An average of 0.06  $\text{kg yr}^{-1}$  PCB emission was estimated for 52 cement production factories (TCMA, 2013). When uncertainty data is included, lower and upper emission levels are likely to be 0.03  $\text{kg yr}^{-1}$  and 0.6  $\text{kg yr}^{-1}$ , respectively. The data for estimating the emissions from smelting processes were obtained from Turkish Foundry Association. In the foundry sector report (TUDOKSAD, 2012) it is stated that off-the-record production in this sector was estimated to be less than 5% of overall production amount. Data for high capacity steel manufacturing industry was obtained from Turkish Steel and Iron Producers Association (TISPA, 2013). A total of 579.3  $\text{kg yr}^{-1}$  of PCBs emission from 31 facilities (electric arc furnaces, electric arc furnaces with preheating and sinter production) was estimated. Two of the plants are equipped with preheated electric arc furnaces. One of them is located in Aliaga while the other one is in Dilovasi. Thus, elevated emissions occurred at those two areas. With the utilization of uncertainty, lower and upper emission levels were estimated to be 290  $\text{kg yr}^{-1}$  and 1 160  $\text{kg yr}^{-1}$ .

Table 2. Activity levels of each industrial sector in Turkey

Production Type	Capacity Classification		
	>20 000 ton yr <sup>-1</sup>	1 000-20 000 ton yr <sup>-1</sup>	<1 000 ton yr <sup>-1</sup>
Pig Iron	18	212	356
Steel	27	35	17
Zinc	4	23	149
Copper	2	7	11
	>2 M tons	1-2 M tons	<1 M ton
Cement	11	22	19
	>1 000 MW	1 000-500 MW	<500 MW
Brown Coal-fired thermal power plant	3	3	5
Hard Coal-fired thermal power plant			1

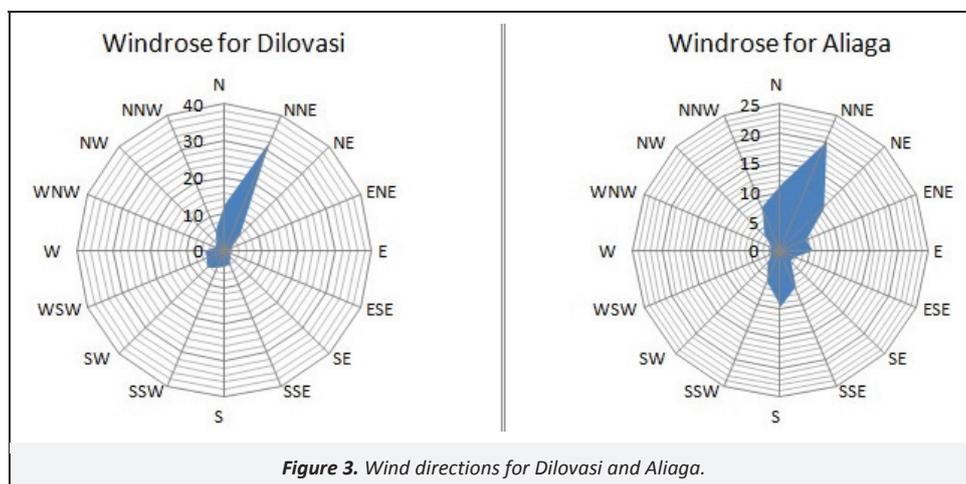


Figure 3. Wind directions for Dilovasi and Aliaga.

There are 12 power plants operating with lignite and hard coal (EUAS, 2012). Only one of them is operating with hard coal. Total annual PCB emission from these thermal power plants was estimated to be 22.03 kg. This value is expected to be between 11 kg yr<sup>-1</sup> and 33.7 kg yr<sup>-1</sup> according to uncertainties. There is only one medical waste incineration plant located in Istanbul. Capacity of the plant is 24 tons day<sup>-1</sup> which corresponds to an estimate of 0.18 kg yr<sup>-1</sup> PCB emission. Lower and upper levels are estimated to be 0.1 kg yr<sup>-1</sup> and 1.8 kg yr<sup>-1</sup>, respectively.

There is one hazardous waste incineration plant in Kocaeli but it was not considered in this study since it has an adsorption unit for the removal of POPs.

Estimated emission results are summarized in Figure 4. Whiskers show lower and upper levels of releases according to uncertainties.

The total PCB emission was found to be 920 kg yr<sup>-1</sup>. Considering the uncertainties, emissions could be as high as 1 738 kg yr<sup>-1</sup> and down to 511 kg yr<sup>-1</sup>. Highest contribution occurs in Kocaeli located in the North–West of the country by 47% of the overall emissions.

Sixty three percent of the releases occur by iron–steel manufacturing activities. Second highest contributor is copper smelting by 33%. Atmospheric releases from cement manufacturing plants and medical waste incineration are negligible when compared to other manufacturing applications.

### 3.2. Spatial distribution of PCB releases over Turkey

Several methods can be employed to prepare a digital pollution map including the Krigging method which is the most commonly used one to interpolate between two sets of spatially distributed data (Casado et al., 1994; Liu and Rossini, 1996; Holland et al., 2000; Kuzu et al., 2009; Oztaner and Sakarya, 2012). Map of Turkey was gridded into 476 grids of 50 km resolution. PCB emission map by Krigging method is shown in Figure 5, in which

the emission scale is in kilograms of PCB per year. The map showed two hotspots, one of which is the Aliaga region in Izmir in the most western part of Turkey. The other hot spot was located around Dilovasi district in Kocaeli. Both of these regions were characterized by heavy industrialization.

### 3.3. Modeling results

Simple box model calculations were utilized for the two detected hot spots. Model input variables and model outputs are listed in Table 3.

Three results were generated for each location. Average results are those which are calculated from emission factors. The other values of minimum and maximum give the concentrations at upper and lower uncertainty levels. Background concentration from the transported air into the box was neglected.

In Aliaga, industrial activities are expected to make a contribution of 126 pg m<sup>-3</sup> on average. When uncertainties are considered, this value can range between 66 pg m<sup>-3</sup> and 246 pg m<sup>-3</sup>. There are several studies in the literature conducted in Aliaga on the quantification of ambient PCB concentrations. Cetin et al. (2007) reported ambient concentrations as 3 136 ± 824 pg m<sup>-3</sup> and 1 371 ± 642 pg m<sup>-3</sup> for summer and winter, respectively. In another study, 3 370 ± 1 617 pg m<sup>-3</sup> and 1 164 ± 618 pg m<sup>-3</sup> were detected in the same location (Bozlakler et al., 2008). Both researchers indicated that concentration increase in the summer months is due to volatilization from contaminated soil. One might think of that model results underestimate ambient concentrations, however it also should be considered that each grid cell of the model is 50 km long. Thus, releases are assumed to be distributed homogenously within the grid and no sink in the grid and no other source is present. If a shorter box length, including iron–steel industries, was chosen, such as 5 km, then the average concentration would be predicted as 1 260 pg m<sup>-3</sup> which is in accordance with the observed results. Major sources in this area could be regarded as the industrial activities in Aliaga.

Table 3. Box model inputs and outputs

	Model Inputs						Model Outputs		
	q (kg yr <sup>-1</sup> )			u (m s <sup>-1</sup> )	H (m)	L (m)	C (pg m <sup>-3</sup> )		
	Min	Avg	Max				Min	Avg	Max
Dilovasi	250	434	800	2.75	410	50 000	140	244	450
Aliaga	150	286	560	3.00	480	50 000	66	126	246

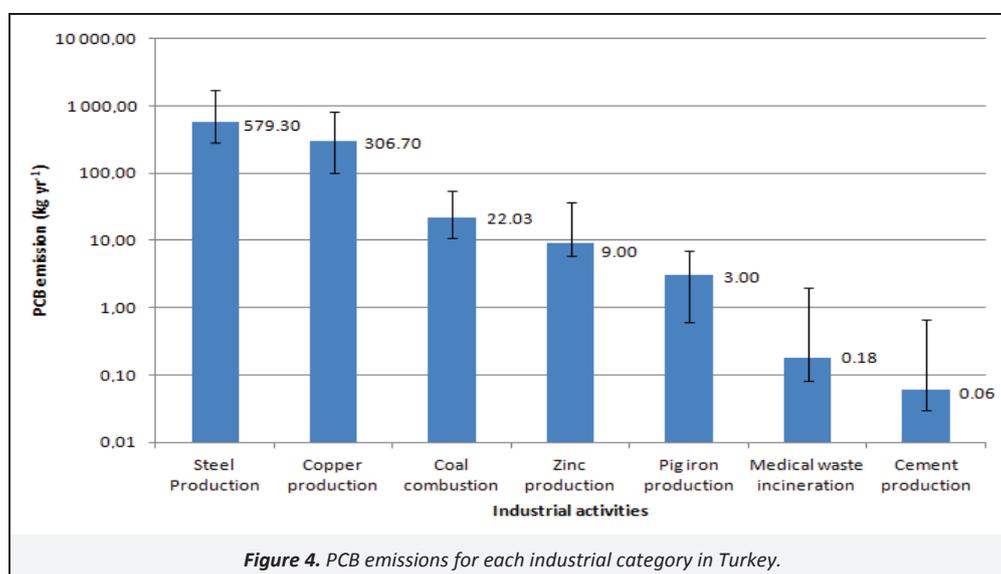
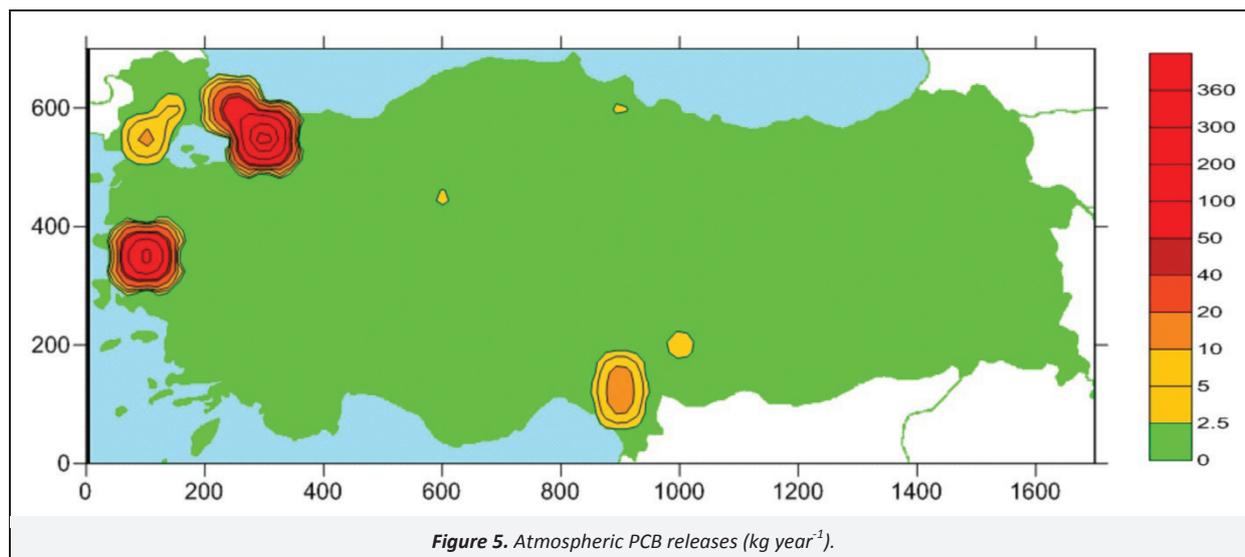


Figure 4. PCB emissions for each industrial category in Turkey.



Model estimates that on the average, ambient concentration contribution in Dilovasi is  $244 \text{ pg m}^{-3}$ . Lower and upper limits are  $140 \text{ pg m}^{-3}$  and  $450 \text{ pg m}^{-3}$ , respectively. Since no ambient air PCB concentration measurements exist in Dilovasi yet, a direct comparison cannot be made as in Aliaga. However, Cindoruk and Tasdemir (2010) reported ambient concentration results from 4 different sites in Bursa located 60 km South West of Dilovasi. Highest ambient concentration was determined to be  $570 \text{ pg m}^{-3}$  in Mudanya near Marmara Sea. As no other significant source located in Mudanya, industrial releases could have been a contributor to ambient concentrations in Mudanya. Transport of PCBs by wet and dry deposition to sea surface, then volatilization by air–water exchange could also be effective as well as direct transport of the pollutants by regional sources. Also, prevailing wind direction from Dilovasi is towards to Mudanya.

#### 4. Conclusions

Recently, a narrow-scoped emission inventory of present PCB load on transformers and capacitors have been prepared for Turkey. But it is indicated in the National Implementation Plan that these results were obtained from questionnaire results and direct measurement of some of the equipments. It could clearly be concluded that, these results doesn't reflect the total actual emissions. In the present study, a different detailed approach was aimed to estimate PCB emissions. Activity levels of PCB emitting industries and their corresponding emission factors were used for the estimation of PCB emissions.

According to estimations highest contribution is from steel making industry. This sector is contributing to the 63% of overall PCB emissions. Second highest release occurred through copper smelting with a contribution of 33%. The remaining sectors release only 6% of the total PCBs. Total PCB emissions throughout the country is approximately  $920 \text{ kg yr}^{-1}$  on the average. When uncertainties are included this value range from  $511 \text{ kg yr}^{-1}$  to  $1738 \text{ kg yr}^{-1}$ .

Two hot spots were located; those are Aliaga and Dilovasi industrial regions. Simple box model applied for these two sites in order to determine their potential importance. Emissions from those sites make contributions of  $126 \text{ pg m}^{-3}$  and  $244 \text{ pg m}^{-3}$  to ambient atmosphere for Aliaga and Dilovasi, respectively. If a shorter grid length is chosen, then higher concentrations are determined. For a 5 km grid length which is more viable at local basis, estimation and observation results for Aliaga were found to be in accordance with the previous studies. Especially, iron–steel manufacturing sector is thought to be the major contributor at

local basis. As a result, it could be suggested that PCB reduction measures from the flue gases must be obligatory in such facilities.

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