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ARAŞTIRMA MAKALESİ **RESEARCH PAPER**

Correlating Gas/Particle-Phase Concentration of Indicator Polychlorinated Biphenyls with Meteorological Factors in Bayburt, an Important University City of Turkey

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Abstract: This study was carried out in two seasons in Bayburt University Baberti Campus (BUBC) in Bayburt city, which is an important university city in Turkey. Gas and particle phase atmospheric concentrations of Urban atmospheric Indicator Polychlorinated Biphenyls (i-PCBs: PCB#28, 52, 101, 118, 138, 153, 180) were determined at first. Then, by using different methods, the interactions between i-PCB concentrations and meteorological factors were investigated. Gas and particle phases of atmospheric i-PCB samples were collected using a high-volume air sampler (HVAS) in BUBC . Following this stage, the samples were analyzed in a gas chromatography-mass spectrophotometer (GC-MS) by passing through appropriate extraction steps. According to the data obtained, the average gas phase ∑i-PCB concentration for the summer month was determined to be 11.314 pg/m^3 during the sampling period while the average particle phase ∑i-PCB concentration was found to be 2.145 pg/m^3. The average gas phase ∑i-PCB concentration was found to be 4.582 pg/m^3 for the winter period while it was found to be 1.756 pg/m^3 for the average particle phase ∑i-PCB concentration. In addition, the average total concentrations of i-PCBs were calculated to be 19.797 pg/m^3. It was determined that meteorological factors especially temperature (from the *Clausius–Clapeyron* equation), wind velocity and direction, and rainfall have significant impacts on i-PCB concentration and its distribution. In light of these data, i-PCB concentrations determined in the gas phase were found to be higher than those in the particle phase. In addition, concentration rates determined in summer months were calculated to be higher compared to those calculated for winter months.

Keywords: Active atmospheric sampling, extraction, GC-MS, i-PCBs, organic pollutants, polychlorinated biphenyls.

Türkiye'nin Önemli Üniversite Kenti Bayburt da İndikatör Poliklorlu Bifeniller'in Gaz/Partikül Faz Konsantrasyonlarının Meteorolojik Faktörler ile İlişkilendirilmesi

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Öz: Bu çalışma, Türkiye'nin önemli Üniversite Kenti olan Bayburt Üniversitesi Baberti Kampüsünde (BÜBK) mevsimsel olarak yapılmıştır. Önce kentsel atmosferik İndikatör Poliklorlu Bifeniller'in (i-PCBs: PCB#28, 52, 101, 118, 138, 153, 180) hem gaz hem de partikül faz atmosferik konsantrasyonları tesbit edilmiştir. Ardından da i-PCB konsantrasyonlarının meteorolojik faktörler ile olan etkileşimleri farklı yöntemler ile araştırılmıştır. BÜBK'de yüksek hacimli hava örnekleleyicisi (YHHÖ) kullanılarak atmosferik i-PCB örneklerinin gaz ve partikül fazları toplanmıştır. Daha sonra uygun ekstraksiyon adımlarından geçirilerek, gaz kromotografi-kütle spektrofotometre'de (GC-MS'de) analizlenmiştir. Elde edilen verilere göre örnekleme periyodu boyunca yaz ayı için ortalama gaz fazı ∑i-PCB konsantrasyonu 11,314 pg/m^3 olarak belirlenirken, ortalama partikül faz ∑i-PCB konsantrasyonu ise 2,145 pg/m^3 olarak tesbit edilmiştir. Diğer yandan kış ayı için de ortalama gaz fazı ∑i-PCB konsantrasyonu 4,582 pg/m^3 olarak belirlenirken, ortalama partikül faz ∑i-PCB konsantrasyonu ise 1,756 pg/m^3 olarak tesbit edilmiştir. Ayrıca i-PCB'lerin ortalama toplam konsantrasyonları (yaz+kış; gaz+partikül faz) 19,797 pg/m^3 olarak hesaplanmıştır. i-PCB konsantrasyon ve dağılımlarına özellikle sıcaklık (*Clausius-Clapeyron eşitliğinden*) başta olmak üzere, rüzgar hızı/yönü ve yağışlar gibi meteorolojik faktörlerin önemli ölçüde etkilediği tesbit edilmiştir. Bu veriler ışığında, tesbit edilen i-PCB türlerine ait i-PCB konsantrasyonları gaz fazındaki değerler, partikül fazından daha yüksek oranda bulunmuştur. Ayrıca yaz aylarında bulunan konsantrasyon değerleri kış aylarına göre daha fazla hesaplanmıştır.

Anahtar Kelimeler: Aktif atmosferik örnekleme, ekstraksiyon, GC-MS, i-PCB'ler, organik kirleticiler, poliklorlu bifeniller.

INTRODUCTION

PCBs are among the persistent organic pollutants (POPs) and have harmful effects on living and non-living environments sheltering humans, animals, and plants. They are precipitated under the effect of meteorological events during their half-life at the time they are released into and suspended in the atmosphere as the result of various human activities (Arar et al., 2019). Therefore, PCBs' atmospheric concentrations and effects on public health have recently been studied extensively in the literature (Chaemfa et al., 2009; Günindi & Tasdemir, 2010; Liu et al., 2019; Trinh et al., 2018).

PCBs are important congeners of POPs and are released fully into the atmosphere as the result of anthropogenic activities. PCBs started to be produced in 1929 for the first time and no study was conducted about their concentrations by 1966 (Bogdal et al., 2013; Combi et al., 2020). In addition, it was forbidden to produce PCBs upon realizing that they have toxic effects (Cabrerizo et al., 2011; Omwoma et al., 2019). However, they still exist in the atmosphere since they were extensively used in previous years and their half-life is long in nature even though their use and production are forbidden all over the world (Baek et al., 2011; van den Dungen et al., 2015; Gregoris et al., 2014; Ullah et al., 2020).

PCBs include each form of $C_{12}H_xCl_y$ (x=0~9, y=10-x) 209 congeners and are carcinogenic (Arar et al., 2019). Among these congeners, 7 PCBs are so-called indicator PCBs (Aslam et al., 2019; Olenycz et al., 2015; Paloluoğlu, 2016). All PCB congeners are considerably stable, non-flammable, soluble in organic solvents, and flammable and explosive compounds (Cindoruk & Taşdemir 2010; Shin et al., 2011; Ullah et al., 2020; Wu et al., 2011). Since PCBs are resistant to degradation in nature, they can easily accumulate in the bodies of living organisms thus resulting in various health and environmental problems (Barbas et al., 2018; Günindi & Taşdemir, 2011; Paloluoğlu, 2016; Tang et al., 2015). Since the PCBs do not come from natural sources their only source is anthropogenic activities. PCBs are released into the atmosphere through the evaporation process from various surfaces such as landfills, open water and soil surfaces, sludge drying beds, and incineration of PCB-containing waste and electronic devices containing PCBs (Bozlaker et al., 2008; Cakıroğulları, 2006; Needham & Ghosh, 2019). PCBs released into the atmosphere are then exposed to photolytic reactions and removed by wet or dry precipitation. PCBs exposed to atmospheric precipitation can be transported to long distances through various meteorological factors thus entering and polluting different atmospheric, soil, and aquatic media (Aslam et al., 2019; Luo et al., 2015; Sharma et al., 2014). In addition, PCBs released into the atmosphere persist remaining by cycling between air-soil and air-water through a continuous evaporation and precipitation process. They are also stable in the atmosphere, can be transported to long distances, and can accumulate in living organisms through the food chain. Due to all the mentioned features above, PCBs are considerably significant for living things (He & Balasubramanian, 2009; Qiu et al., 2020; Vecchiato et al., 2015; Vijaya Bhaskar Reddy et al., 2019).

It is the first in the study to use an active sampler to investigate atmospheric i-PCBs in BUBC, an urban area in Bayburt. In addition, i-PCB concentration rates tried to be associated with meteorological factors measured at the date of sampling. Totally 7 congeners of i-PCB were sampled and analyzed seasonally for 2 weeks in both summer and winter. In addition, in the present study, gas and particle phase concentrations of atmospheric i-PCBs were determined in the urban atmosphere of Bayburt to find a serious database for the subject. It was also investigated whether the meteorological factors (temperature, precipitation, wind, and wind direction) affect the formation sources of the PCBs. In this regard, it was investigated how meteorological factors especially temperature (from the *Clausius-Clapeyron* equation), wind direction/velocity (from the wind direction and velocity graphs), and precipitation (from precipitation graphs) affect i-PCB concentration and distribution using different methods. This is the first study in the literature to sample and analyze i-PCBs for Bayburt City.

MATERIAL AND METHOD

Sampling Points: Bayburt is an important winter tourism and university city with nearly 15.000 students in the Northeast Anatolia Region. The population of the city center is nearly 80.000 inhabitants. Since the city harbors no significant industrial facilities, nearly no PCB pollutant input is thought to be present in the city's atmosphere. However, due to the geographical and climatic features of the city, serious air pollution problem is experienced resulting from domestic heating and traffic emissions (Kılıç, 2008). The sampling point is located in the university campus (BUBC) in the city center (Figure 1), where it is affected by the city atmosphere in terms of PCB source.

In the scope of the study, the Techora brand active sampler was used to investigate urban atmospheric i-PCBs between 01 and 15 July 2021 and 01 and 15 January 2022 as 2-week active winter and summer samplings respectively. The sampling was performed in BUBC in the city center of Bayburt. Samples were taken through an active Hi-Vol sampler (HVAS) in the air flux rates of 320 m^3/day. While atmospheric gas phase i-PCB samples were collected through Polyurethane Foams (PUF). Glass Fibre Filters

(GFF) were used for particle (PM) phase i-PCBs. In addition, the first PUF cartridges used to determine the gas phase concentrations of the i-PCBs and the GFFs used to determine the particle phase were placed in the active HVAS on 01th July 2021 and 01th January 2022 in summer and winter, respectively. The dirty PUFs and GFFs were replaced with the clean ones in 24-hour intervals. At the end of the summer and winter study periods, the last PUF and GFF were taken from the sampler on 15th July 2021 and 15th January 2022 for summer and winter respectively. Hence, a total of 30 PUF and 30 GFF (summer + winter) were taken at the sampling point. Figure 1 gives seasonal images of the sampling point and system.

Before the sampling procedure, the preparation of PUF cartridges and GFF filters for sampling was performed in convenience with those described in Paloluoğlu, (2016). In addition, the Davis Vantage Pro2 model meteorology station was used at the sampling point to record meteorological parameters (daily temperature, pressure, wind direction, precipitation, humidity, pressure) (Tables 1 and 2). Sampling dates and summer sampling with meteorological parameters (between 01st July 2021 and 15th July 2021) and winter sampling dates with meteorological data (between 01st January 2022 and 15th January 2022) obtained are shown in Table 1 and 2 respectively.

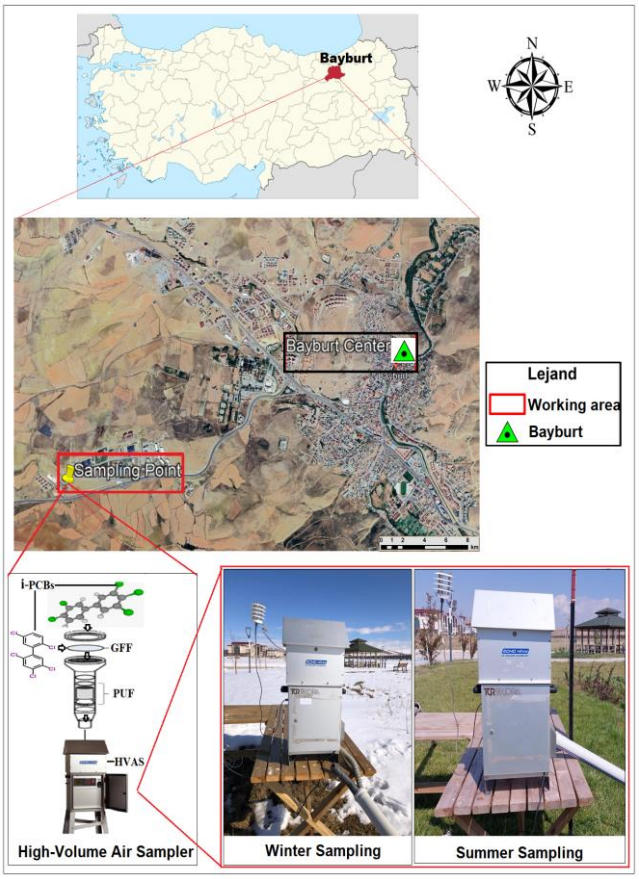


Figure 1: Sampling point.

Table 1. Meteorological data for the summer sampling period.

Dates	Prevalent wind direction	Daily maximum wind speed (at 10m; m/s)	Daily mean wind speed (m/s)	Temperature (ºC)	Daily total precipitation amount (mm)
01.07.2021	ENE	13.0	1.7	31.5	4
02.07.2021	W	13.1	1.3	29.0	0.1
03.07.2021	NE	11.0	1.6	29.1	0
04.07.2021	SSW	14.5	2.1	29.9	0
05.07.2021	W	13.6	1.7	25.3	0
06.07.2021	ENE	10.0	2.6	25.7	4.2
07.07.2021	SSE	9.00	1.2	26.3	14.0
08.07.2021	NE	10.0	2.1	25.0	0.5
09.07.2021	E	14.	2.2	29.5	0
10.07.2021	ENE	9.00	1.6	32.0	0.1
11.07.2021	E	7.50	0.8	32.5	0
12.07.2021	SSW	11.5	0.9	33.5	0
13.07.2021	S	13.5	1.5	30.5	0
14.07.2021	NE	14.5	1.7	28.5	0.3
15.07.2021	NW	15.0	2.3	27.5	13.5

Table 2 Meteorological data for the winter sampling period.

Dates	Prevalent wind direction	Daily maximum wind speed (at 10m; m/s)	Daily mean wind speed (m/s)	Temperature (ºC)	Daily total precipitation amount (mm)
01.01.2022	SW	3.05	0.3	-11.0	0
02.01.2022	S	14.0	1.6	-14.5	0
03.01.2022	WSW	7.50	0.9	-19.0	0.1
04.01.2022	W	14.1	1.2	-10.0	0
05.01.2022	WSW	14.5	2.5	-3.30	0
06.01.2022	W	8.40	1.4	-6.90	9.9
07.01.2022	WNW	11.0	0.8	-18.8	4.5
08.01.2022	WSW	7.50	0.8	-12.9	0.5
09.01.2022	W	9.00	1.1	-7.00	2.1
10.01.2022	SW	12.0	1.9	-19.8	6.5
11.01.2022	SW	12.8	0.9	-12.5	0
12.01.2022	ENE	13.2	2.2	-3.50	0
13.01.2022	E	10.5	1.6	-4.90	0
14.01.2022	W	5.00	0.7	-7.00	0.3
15.01.2022	ENE	6.50	3.1	-8.00	0.1

Extraction and Analysis: Following the sampling operation, particle i-PCB filters (GFFs) were extracted using DCM-PE (Dichloromethane-Petroleum ether) in an Ultrasonic bath. Gas phase PCB filters were extracted using a mixture of (PUF) ACE-HEX-DCM (Acetone-Hexane-Dichloride Methane) in the Soxhlet extractor (Figure 2). Gas and particle phase extracted atmospheric i-PCB samples were analyzed using a GC-MS (Gas Chromatography-Mass Spectrometer; Agilent brand) device (Paloluoğlu et al., 2016). GC-MS study parameters were selected according to Cindoruk and Taşdemir, (2010). A tight quality control procedure was applied in all applications.

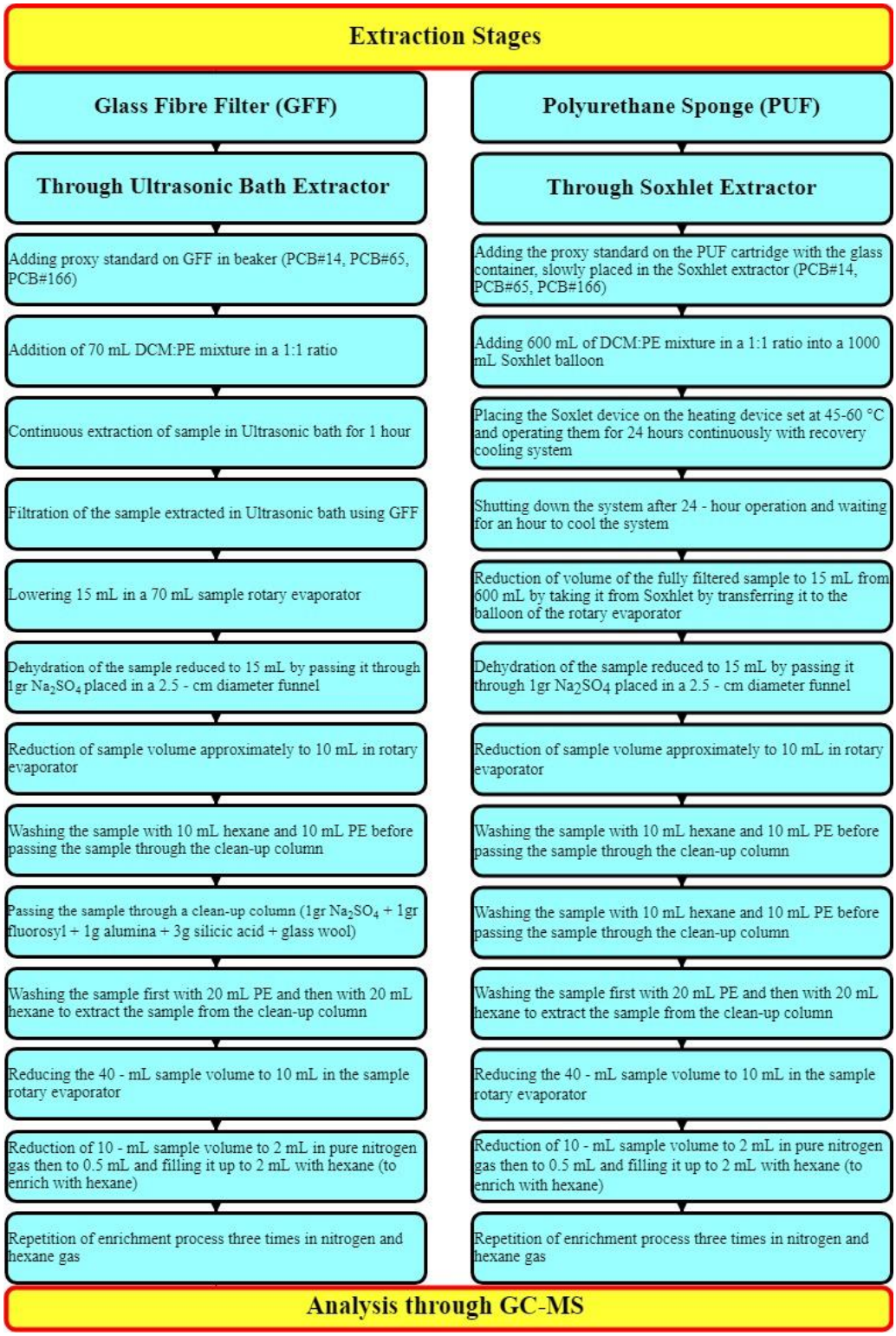


Figure 2. Extraction stages of Atmospheric i-PCBs.

Quality Control and Quality Reliability: Widely accepted studies in international literature were utilized for the accurateness of the results of sampling, extraction, and analyses (Barbas et al., 2018; Cindoruk & Taşdemir, 2010; Paloluoğlu, 2008; Ullah et al., 2020; Bhaskar Reddy et al., 2019). In this respect, a serious procedure was adopted for Vijaya quality control and reliability at all stages of the study. *Calibration standards* of atmospheric indicator PCBs were investigated for 7 i-PCB congeners (PCB#28, PCB#52, PCB#101, PCB#118, PCB#153, PCB#138, PCB#180). Six different concentration rates in 1-µL injections were realized in GC-MS analysis for calibration in hexane standard solution. *Surrogate standards* were also used to consider the losses to be seen in the analysis. PCB#14 (3,5-diklorobifenil), PCB#65 (2,3,5,6-tetraklorobifenil) and PCB#166 (2,3,4,4'5,6-hekzaklorobifenil) were added in hexane solution in the rate of 5 ppb (ng / mL). In addition, PCB#14, PCB#65, and PCB#166 surrogate standards were categorized to calculate the analytic recovery of 7 i-PCB congeners. According to this categorization, ***PCB#14*** was attributed to PCB#28 congener, ***PCB#65*** to PCB#52, PCB#101, PCB#118, PCB#153, PCB#138 congeners, and ***PCB#166*** to PCB#180 congener. Table 3 gives the highest and lowest recovery rates (standard percentage) obtained in the study.

Table 3. Recovery percentages of i-PCBs taken sampling (%).

Surrogate Standards	HVAS (GFF)	HVAS (PUF)	Mean
PCB#14	80	69	75
PCB#65	95	88	89
PCB#166	101	99	103

PCB#30 (2,4,6 trichlorobiphenyl) and PCB#204 (2,2',3,4,4',5,6,6') congeners were used as volume removal (*internal standards*) in the study. Atmospheric i-PCB samples taken through the *Internal standards* (volume adjustment) were used for volume adjustment before being taken to the GC-MS device. These internal standards were added in each bottle at the rate of 1ppb (ng/mL) concentration before GC-MS injection. Volume adjustment was performed considering the results obtained. *Instrument Detection Limit (IDL)* was determined through the method used in Cindoruk and Tasdemir, (2007). The lowest and highest instrument detection limit value (IDL) ranges of the i-PCB congeners according to the sample type group were calculated as 0-0.81 ng for PUF and 0-0.85 ng for GFF. In addition, 4 field and 2 laboratory blanks were taken at the sampling point. The ratio of mean PCB rates measured in blank samples to total PCB rates measured in atmospheric indicator samples was found to be 0.2% for PUF and 4.3% for GFF. In this way, percentages of PCB pollution input were calculated. Recovery percentages were also taken into account. Atmospheric i-PCB exact values were calculated by adding or distracting the result concentration values.

RESULTS AND DISCUSSION

In the study conducted for the first time in the northeast of Turkey, summer and winter season changes of atmospheric PCBs were monitored for 7 i-PCB congeners in an urban (BUBC) area. A total of two 15-day active atmospheric samplings were performed in the summer and winter seasons on atmospheric gas and particle phase i-PCBs. In summer and winter sampling, all of the 7 i-PCB congeners were found in gas and particle phases. During the summer sampling period, the mean gas phase ∑i-PCB concentration was measured to be 11.314 pg/m^3. In the same way, during the summer sampling period, the mean particle phase ∑i-PCB concentration was measured to be 2.145 pg/m^3. Likewise, the average gas phase ∑i-PCB concentration was measured as 4.582 pg/m^3 during the winter sampling period. The particle phase mean ∑i-PCB concentration was also calculated as 1.756 pg/m^3. Mean gas and particle phase concentrations of atmospheric i-PCBs (pg/m^3) obtained in summer and winter sampling periods are given in Table 4.

Table 4. Mean gas and particle phase concentrations of atmospheric i-PCBs (pg/m^3) obtained in summer and winter sampling periods (pg/m^3).

	BUBC Sampling Area / Atmospheric i-PCB Concentrations							
	1st – 15th July 2021 Summer sampling				**1st – 15th January 2022 Winter sampling**			
	Gas phase		**Particle phase**		**Gas phase**		**Particle phase**	
i-PCBs	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**
Tri-PCBs								
PCB#28	6.090	5.900	1.200	1.155	1.990	1.900	0.811	0.690
Tetra-PCBs								
PCB#52	3.995	3.451	0.650	0.570	1.910	1.850	0.650	0.605
Penta-PCBs								
PCB#101	0.483	0.535	0.065	0.061	0.068	0.064	0.066	0.060
PCB#118	0.107	0.070	0.054	0.050	0.064	0.055	0.052	0.048
Hexa-PCBs								
PCB#153	0.475	0.432	0.064	0.062	0.072	0.069	0.057	0.052
PCB#138	0.084	0.050	0.060	0.053	0.058	0.050	0.060	0.055
Hepta-PCBs								
PCB#180	0.080	0.070	0.052	0.049	0.420	0.399	0.060	0.054
∑7i-PCBs	**11.314**		**2.145**		**4.582**		**1.756**	

*** Mean =** Results represent average, **SD =** Standard deviation; **∑7i-PCBs** = Sum of average concentrations of PCB#28, PCB#52, PCB#101, PCB#118, PCB#153, PCB#138, PCB#180

As can be seen from Tables 4 and 5, the concentration values of the i-PCBs determined seasonally were found to be higher in summer compared to winter. In addition, the gas phase concentration values of i-PCBs were also found to be higher than those in the particle phase. Such results are similar to the results of several studies in the literature (Cetin et al., 2017; Cindoruk & Taşdemir, 2010; Chakraborty et al., 2013; Trinh et al., 2018; Wang et al., 2016).

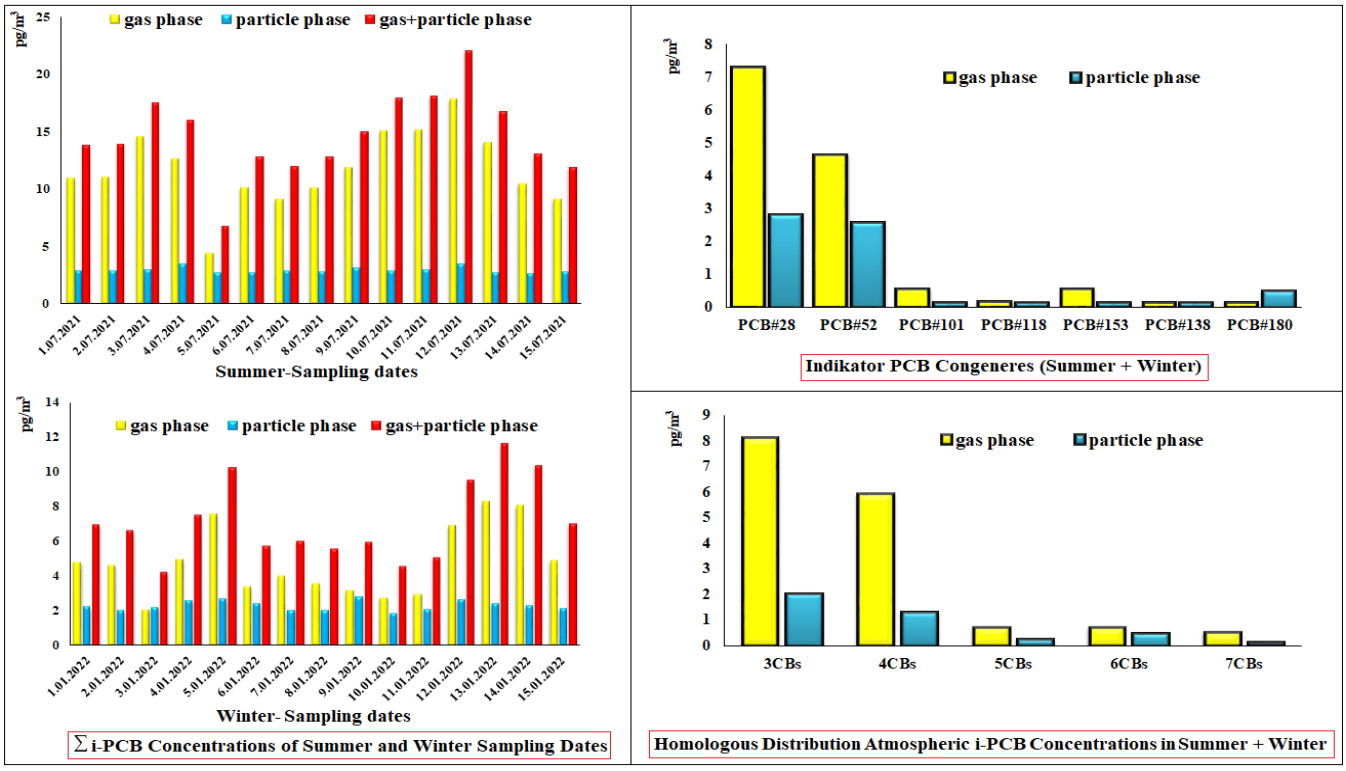
Table 5. Mean gas and particle phase concentrations of atmospheric i-PCBs for summer and winter periods (pg/m^3) and presence percentages (%).

i-PCBs	BUBC Sampling Area			
	1st–15th July 2021 Summer sampling		1st– 15thJanuary 2022 Winter sampling	
	Cpg/m^3	%	Cpg/m^3	%
Tri-PCBs				
PCB#28	**7.290**	**54.16**	**2.801**	**44.19**
Tetra-PCBs				
PCB#52	4.645	34.51	2.560	40.40
Penta-PCBs				
PCB#101	0.548	4.07	0.134	2.11
PCB#118	0.161	1.20	0.116	1.83
Hexa-PCBs				
PCB#153	0.539	4.01	0.129	2.04
PCB#138	0.144	1.07	0.118	1.86
Hepta-PCBs				
PCB#180	0.132	0.98	0.480	7.57
∑7i-PCBs	**13.459**		**6.338**	

*∑**7i-PCBs** = Sum of average concentrations of PCB#28, PCB#52, PCB#101, PCB#118, PCB#153, PCB#138, PCB#180; **Cpg/m^3** = gas + particle phase sum

In the seasonal sampling period, the most dominant atmospheric indicator PCB congeners were determined to be PCB#28 and PCB#52 in the gas and particle phase. In addition, other 5 i-PCB congeners were also determined to be present. It was determined in the active atmosphere sampling that the highest concentration values for summer and winter months belong to PCB#28 in the gas and particle phase with 7.209 pg/m^3 and 2.801 pg/m^3, respectively. Similar studies in literature give similar results where among atmospheric i-PCBs, PCB#28 is the most dominant congener (Bogdal et al., 2013; Frederiksen et al., 2012; Wu et al., 2018).

Atmospheric concentrations of 7 i-PCB congeners were found to be PCB#28, 52, 101, 118, 153, 138, and 180 for summer + winter periods and gas and particle phases (Figure 3). Among these congeners, PCB#28 was detected at higher levels than other indicator congeners in the BUBC (Figure 3). PCB congeners with mostly low chlorine content (3- and 4-CBs) were detected in higher concentrations compared to other less volatile (5-, 6- and 7-CBs) congeners (Arinaitwe et al., 2018; Cetin et al., 2017; Gevao et al., 2017; Yang et al., 2012).



Colorings were used to show the gas and particle phase concentration differences of i-PCBs (Yellow color: gas phase, Blue color: particle phase, and Red color: gas + particle phase).

Figure 3: Atmospheric indicator PCB concentrations (pg/m^3).

As can be seen in Figure 3, it is thought that PCB#101, PCB#118, PCB#153, PCB#138 and PCB#180 congeners were detected to be at low concentrations since they are in the high chlorinated biphenyl group and have insufficient resources (Afful et al., 2013; Mahmood et al., 2014; Ali et al., 2015; Cetin et al., 2018). The changes in the total concentrations of the gas phase, particle phase, and gas + particle phases of the i-PCB congeners detected in the BUBC point are given in (Figure 3).

It is seen when Figure 3 is evaluated that atmospheric i-PCB concentrations show daily differences. The highest i-PCB concentration was determined on the

sampling date of 12th July 2021, when the temperature was the highest (Table 1: 33 °C). It is also seen from (Table 1) that the daily mean wind speed was lower (0.9 m/s) and no rainfall was recorded (0 mm). Based on this observation it is seen that PCB concentrations are directly proportional to temperature and inversely proportional to wind speed and precipitation. On the sampling day of 05th July 2021, the concentration rate of i-PCB was the lowest wind speed was high with 1.8 m/s and the temperature was lower (26.3 °C; Table 1). On the day when temperature was lower and wind speed was higher, i-PCB concentrations were also lower and such a situation can reflect the important role of meteorological factors in changing i-PCB concentrations.

It is seen from the column graphs in Figure 3 that the winter period total gas + particle phase i-PCB concentrations face a wider range of fluctuation than in the summer period. When the effect of meteorological factors on i-PCBs in the winter period is examined, it is determined that the i-PCB concentrations are the highest on the days 05, 13, 14.01.2022, especially on dry days. It was also determined that the i-PCB concentrations were lower on wet days with precipitation amounts between 0.1 and 9.9 mm (Table 2). The data obtained suggests that i-PCB concentrations may change inversely with precipitation. Similarly, it was observed during the winter period that i-PCB concentrations changed inversely to wind speed (Table 2).

The homologous distribution of gas and particle phase atmospheric i-PCB concentrations in summer and winter in the BUBC area is presented in Figure 3. PCB#28 and PCB#52 from 3- and 4-CB indicator groups with low molecular weight were found to be dominant in summer and winter. In addition, 3-CBs represented the larger values at the rate of 54% in both particle and gas phases than other indicator homologous groups. Other indicator homologous groups following this group are PCB#52 (34.5%) from 4-CBs, PCB#101 from 5-CBs and 118 (5.3%), PCB#153 and 138 from 6-CBs (5.1%), and PCB#180 (0,98%) from 7-CBs,

In the studies in literature, the most dominant homologous groups were determined to be 3-CBs (PCB#17, ***18***, 31, 26, 19, 22, ***28***, 32, 37) and 4-CBs (PCB#44, ***52***, 49, 47, 70, 64, 71, 66, 74, 77) atmospheric active sampling conducted in urban, semi-rural and rural areas as in the present study. PCB#28 and PCB#52 in 3- and 4-CBs were found to be more dominant in the atmospheric environment compared to other homologous groups. The most apparent factors of this situation are the fact that the molecular weight of PCB#28 and PCB#52 is low and they are used densely in the *Aroclor* group with a low number (Cindoruk & Taşdemir, 2010; Giuliani et al., 2015; Wu et al., 2018).

Because the city of Bayburt is less developed compared to Western countries in terms of heavy industry, industrial facilities, and population, it is expected to be quite weak in terms of atmospheric i-PCB source. However, according to the results obtained, detecting the higher rate of low-chlorinated biphenyls can correspond volatile properties of PCBs and their transportation to different regions by atmospheric movement.

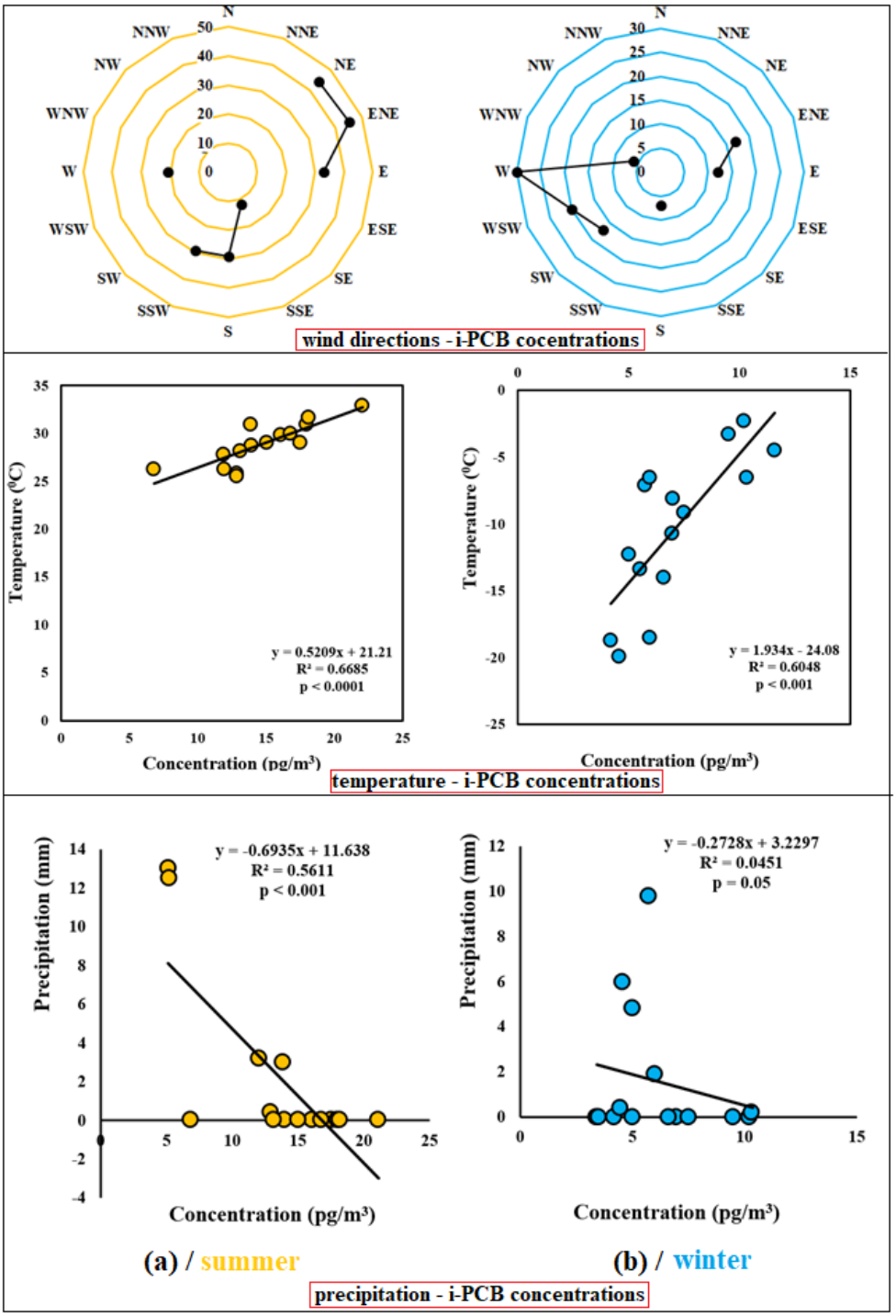
The results obtained in this study are similar to other results in the literature in terms of homologous groups. In a similar study in literature Manodori et al., (2007) examined 54 PCB congeners in atmospheric active sampling between 2002-March and 2003-June in Kyonggi-do city, North Korea, and determined the most dominant homologous groups as 3-, 4, 5-, 6-, 7-CBs. In another study, Cindoruk & Taşdemir, (2010) examined totally 41 PCB congeners in atmospheric active sampling between August 2004 and April 2005 in Bursa, Turkey, and determined 3-, 4- and 5-CB to be the dominant homologous group. In the study of Barbas et al., (2018), a total of 68 PCB congeners were examined in the atmospheric active sampling conducted between January and December 2013 in Madrid, Spain, and determined the dominant homologous group as 3-, 4-, 5-, 6-CB. Liu et al., (2020) examined 19 PCB congeners in their sampling in Taizhou, China, between December 2016 and October 2017, and they determined 2-, 3-, and 4-CB as the dominant homologous group. As can be seen from similar studies, in the present study, the most dominant homologous groups were determined as 3-CB and 4-CBs.

Relationship Between Atmospheric I-PCBs and Meteorological Parameters: Among meteorological parameters, precipitation (snow, rain), temperature wind speed, and direction play an important role in air quality and organic pollutants. Temperature, one of the most important factors, can result in the transportation of pollutants (organic pollutants) from surfaces (soil, snow, vegetation, etc.) by evaporating over short or long distances. In addition, precipitation can clean the atmosphere, and transportation and dilution of pollutants in the environment can be possible through wind (Cindoruk & Taşdemir, 2010; Hussain et al., 2019 Wang et al., 2017; Yurdakul et al., 2019). In this respect, the effects of some meteorological parameters on the total atmospheric i-PCB concentrations obtained from the study area are compared below.

***Effect of Wind Direction and Velocity on Active Atmospheric I-PCB Concentrations*:** The distribution of PCBs according to prevalent wind directions was measured by using the prevalent wind directions in BUBC and the average concentrations of total gas + particle belonging to the area and results are given in Figure 4a (Summer) and b (Winter). In the summer sampling period, measured wind

directions were south-southwest (SSW), south (S) east-northeast (ENE), and east (E). The highest concentration values of atmospheric i-PCBs were observed on the days when wind blew from the directions above. Since small industrial facilities and residential areas are located in the south-south-west directions of the BUBC area, it is possible that PCB pollutant inputs can be transported from this direction. In addition, in the BUBC area, the highest atmospheric i-PCB concentrations in the winter sampling period were measured on days when the wind blew from the west (W) south-west (SW), and east-north-east (ENE) directions (Figure 4b).

The Solid Waste Facility of Bayburt is located in ENE of the BUBC area while small industrial facilities, where in winter PCB-containing materials are burned and emitted into the atmosphere, are located in the SW as well as a densely populated residential area. Such a situation in the area increases the possibility of air movement from this direction containing i-PCB.



*Colorings were used to show seasonal differences (Yellow color: Summer season, Blue color: Winter season).

Figure 4: Variation of i-PCB concentrations with meteorological factors.

Effect of Temperature on Active Atmospheric I-PCB Concentrations: Temperature is an important meteorological factor affecting atmospheric i-PCB concentrations. It was determined in several studies in the literature that i-PCBs can be transferred into the atmosphere as the result of evaporation from various surfaces on earth (like waste storage, soil, water surfaces, and snow cover) with the effect of temperature (Aslam et al., 2019; Bozlaker et al., 2008; Sharma et al., 2014; Ali et al., 2015). Concentrations of atmospheric i-PCBs were found to be at the highest level at sampling time, especially on the hottest days of July. The effect of temperature was observed clearly on i-PCB congeners in especially summer time (R^2= 0.67; p<0.0001) (Figure 4a) (Castro-Jimenez et al., 2009; Garcia & Perez, 2003; Yurdakul et al., 2019).

Effect of Precipitation on Active Atmospheric I-PCB Concentrations: Several authors suggested that atmospheric i-PCB concentrations are affected by precipitation (snow, rain, etc.) significantly (Bozlaker et al., 2008; Günindi & Taşdemir, 2010; Harrad & Mao, 2004; Hermanson et al., 2020). In 30-day winter (15 days) and summer (15 days) measurement periods, the number of wet days in summer is only 8 in the sampling area. Total precipitation is 36.7 mm. In winter, the total amount of precipitation is 24 mm over an 8-day snowy period. It is thought that rainfall plays an important role in the measurement of lower rates of atmospheric i-PCB concentrations in especially winter months. Graphics showing the changes in atmospheric i-PCB concentrations with precipitation are given in Figure 4a for summer and 4b for winter. In addition, in January when snowfall is heavy, and in July when the temperature is the highest as well as on windy and wet days, atmospheric i-PCB concentrations were measured to be the lowest (Tables 1 and 2). As can be seen in Figures 4a and 4b, although consistent results are not obtained in the winter months, it shows how important the precipitation for the summer months is in the change of i-PCB concentrations (summer: R^2=0.66; p<0.001 - winter: R^2=0.045; p=0.05).

***Determination of The Relationship Between Clausius-Clapeyron Gas Phase Atmospheric I-PCB Concentrations and Air Temperature*:** *Clausius-Clapeyron* equation expresses that with the effect of evaporation concentration of volatile or semi-volatile organic compounds in gas phase can increase (Eq. 1) (Barbas et al., 2018; Esen, 2006; Sofuoğlu et al., 2001; Yeo et al., 2004).

$$\boldsymbol{LnPa} = m\,(1/T) + b \qquad \textbf{(1)}$$

According to Equation 1, the partial pressure of the gas phase is represented as LnPa (atmospheric partial pressure). Air temperature (T; Kelvin = ^{0}K) is converted into mathematical formulae in inverse proportion to this concentration. The values of slope in Equation 1 (m) and cross point (b) are used to obtain a linear relationship (Barbas et al., 2018; Cindoruk, 2007; Lohmann et al., 2000). In this study, *Clausius-Clapeyron* curves represent BUBC sampling area in summer and winter using data of 7 i-PCB components. When curves belonging to BUBC point are evaluated *Clausius-Clapeyron* graphic slope values are calculated to be -11742 (PCB#28), -11786 (PCB#52), -13652 (PCB#101), -2479.10 (PCB#118), -889.88(PCB#138), -2018.70 (PCB#153), -1463.10 (PCB#180) and -10075 (∑i-PCB) for summer (Figure 5). According to these negative correlations, a very significant relationship was determined between PCB#118 (R^2=0.70; p<0.0001), PCB 28 (R^2=0.53; p<0.001), and PCB#52 (R^2 = 0.41; p<0.01) gas phase PCB concentrations (Figure 5). The last relationship was determined between temperature and summer i-PCB component PCB#180 (R^2 = 0.163; p=0.135). PCB#180 is in the 7-CB group and its molecular weight is one of the highest therefore its rate is thought to be low in the atmosphere inversely proportional to temperature. In one of the similar studies in literature, Yeo et al., (2004) determined in the study conducted in an urban environment that PCB#28 (R^2=0.79; p < 0.0001) and PCB#52 (R^2=0.44; p < 0.01) indicator components have negative correlations with temperature close to the values in the present study by stating that temperature is an important factor. Similar values and results were also obtained from the study of Barbas et al., (2018) involving urban atmospheric PCBs, where i-PCBs, PCB#28 (R^2=0.57; p<0.001), PCB#52 (R^2=0.62; p<0.01) and PCB#101 (R^2=0.47; p<0.05) were found to have a strong relationship with temperature (from LnPa – 1/T graphics) by calculating R^2 values.

As can be seen from the curves in the graphics of the summer period LnPa - 1/T, the temperature is very important in the evaporation from the surfaces and sources (soil, water bodies, PCB-containing materials, and storage areas) and the increase and decrease of the rate of i-PCBs (Figure 5: Currado & Harrad, 2000; Simcik et al., 1999; Cindoruk & Taşdemir, 2007). There are several studies in the literature supporting this finding. Among such studies, that conducted by Cindoruk & Taşdemir (2007) on urban atmosphere calculated the r^2 = 0.82 and p<0.001 values obtained from the LnPa - 1/T graph in a total of 37 PCB components together with i-PCBs. It was found that temperature is a very important factor in PCBs. Likewise, Yeo et al., (2004) examined 11 types of PCB components together with the i-PCBs they researched. They calculated R^2=0.75 and p<0.0001 values that they obtained from the LnPa - 1/T graph quite consistently. Thus, they demonstrated once again what an important effect temperature has on PCBs.

In other similar studies in literature conducted in urban areas, the sources of atmospheric i-PCBs are detected depending on the sloppiness of curves in the LnPa - 1/T graphics. The high sloppiness of the lines on the *Clausius-Clapeyron* graphs shows that the source of total i-PCBs is influenced by regional sources more than air movements and transport (Barbas et al., 2018; Cindoruk & Taşdemir, 2007; Yeo et al., 2004). If the sloppiness is low it may explain that the detected atmospheric i-PCBs are transported to short, moderate, or long ranges (Barbas et al., 2018; Cindoruk & Taşdemir, 2007; Sofuoğlu et al., 2001; Wania et al., 1998; Yeo et al., 2004).

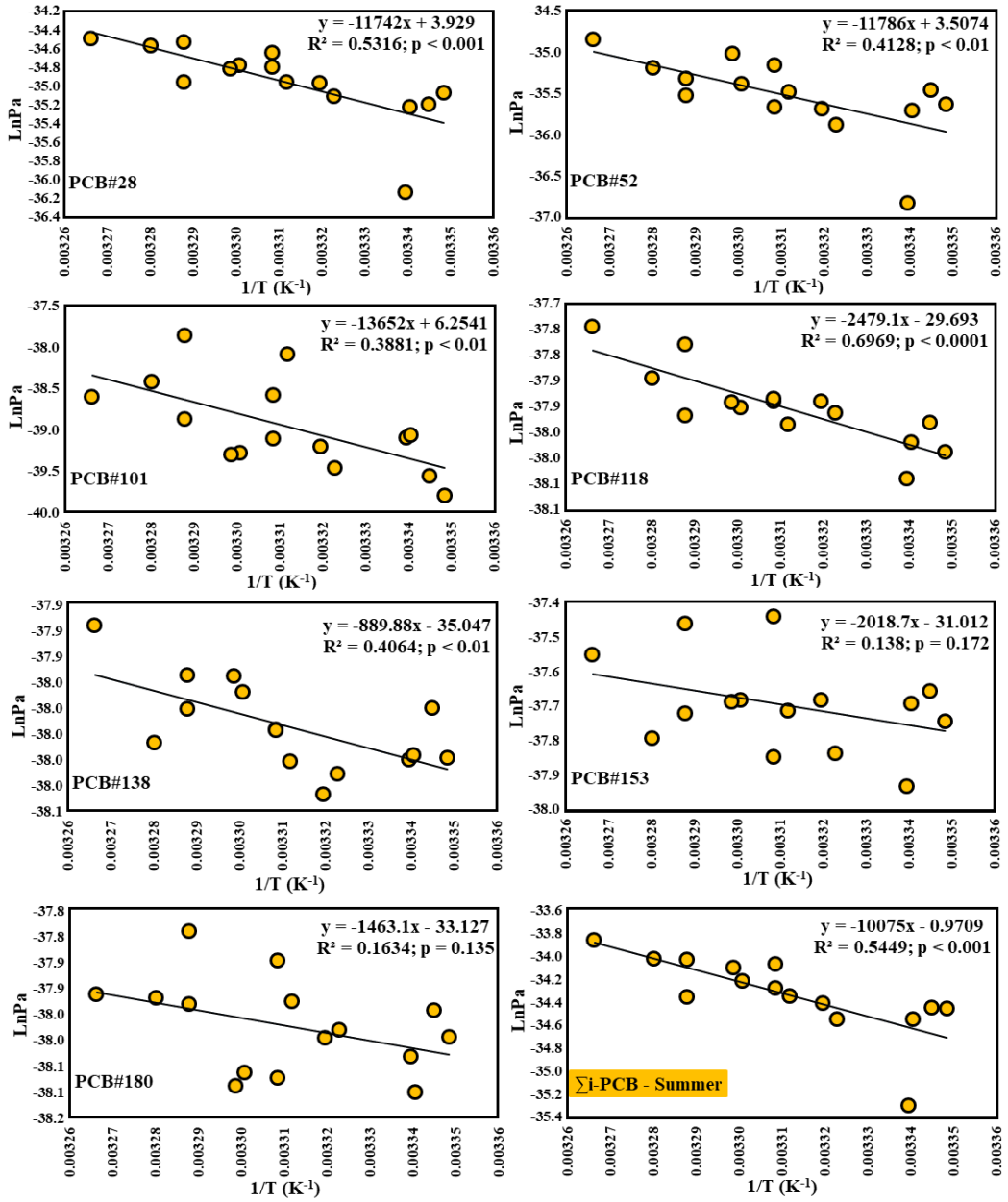


Figure 5: *Clausius-Clapeyron* graphics of atmospheric i-PCB belonging to the summer season.

BUBC area shows similar features for total i-PCBs in summer and winter but each i-PCB represents different features according to seasons. In winter time, each i-PCB curve value was calculated to be -6503.20 (PCB#28), -4741.30 (PCB#52), -520.76 (PCB#101), 836.63 (PCB#118), -54.27 (PCB#138), +25.41 (PCB#153), -2155.30 (PCB#180) and total value was -3952.40 (∑i-PCB: Figure 6). According to negative and positive correlation values obtained, there is an inconsistent relationship between gas phase PCB concentrations and air temperature in the BUBC area in winter time for each i-PCB. It was determined that there is a consistent relationship among a total of 7 i-PCBs (Figure 6). In similar studies in literature, *Clausius-Clapeyron* for PCBs (LnPa - 1/T) curve values are generally negative in the graphics distribution (Cindoruk & Taşdemir, 2007 = -3782.3; Barbas et al., 2018 = -4989 / -9998.9; Harrad & Mao, 2004 = -2800 / -13500; Yeo et al., 2004 = -1490 / -

5673). In the present study, similar negative values were obtained. In again literature, in LnPa – 1/T graphics distributions curve values show a positive correlation (Yeo et al., 2004 = +289 (PCB#60); +2737 (PCB#61/74). In the present study, in the winter time, among i-PCBs, the PCB#118 (+836.63) component has a positive curve value. When literature is reviewed for winter studies low curve values in LnPa - 1/T graphics are seen to be related to atmospheric air movements of pollutants in short, moderate, and long distances (Barbas et al., 2018; Manodori et al., 2007). Higher curve values are based on more regional PCB-containing resources (Harrad & Mao, 2004; Sofuoğlu et al., 2001; Yeo et al., 2003; Yeo et al., 2004). According to the results in the literature, in BUBC sampling points especially in summer months based on high curve angled correlations regional resources can evaporate depending on temperature and carry i-PCB pollution into the atmosphere. It is understood that in winter as the result of lower curve correlation analysis i-PCB pollution input is formed due to short, moderate, and long-range transports. The range of curve values obtained in the present study is in the range of those presented in the literature. For instance, Cindoruk and Taşdemir, (2007) in their study conducted in the urban area found the curve value of the relationship between gas phase PCB concentrations and air temperature to be -3782. In another study by Barbas et al., (2018), the curve value correlating gas phase PCB concentration and the air temperature was found to be between -4989 and -9998.9. In the study of Harrad and Mao, (2004) conducted for urban areas, curve values correlating gas phase PCB concentrations and air temperature were found to be between -2800 and -13500. The negative correlation of total and individual i-PCBs is given in Figures 5 (summer) and 6 (winter).

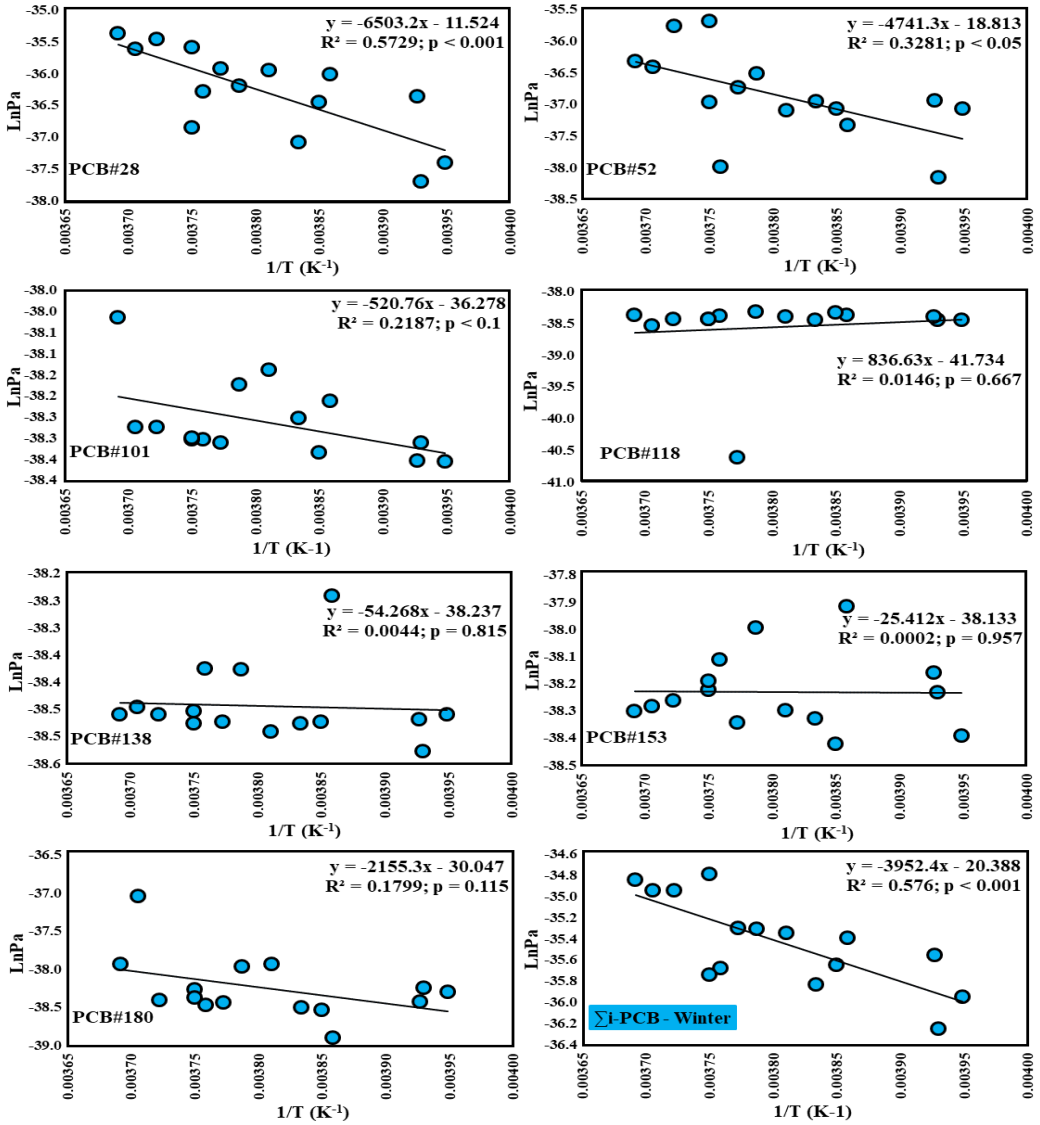


Figure 6: *Clausius-Clapeyron* graphics of winter atmospheric indicator PCBs.

CONCLUSION

Changes in 7 i-PCB components were investigated in an urban environment, BUBC, in the study, which is the first to be conducted in the northeast part of Turkey. It was determined that the mean total (gas + particle) atmospheric i-PCB concentrations measured in the BUBC sampling station changed between 0.26 and 10.01 pg/m^3. In addition, the mean total gas and particle phase values of summer and winter measurements of atmospheric i-PCB components are 19.797 pg/m^3. It is observed that the gas phase of atmospheric i-PCBs is the dominant component phase while PCB homologous groups with low chlorine numbers were also determined to be dominant. The majority of i-PCB congeners were measured to be 3-CBs. The finding that low-CBs are dominant in the area suggests long and short-range transports. The relation between i-PCB concentrations and meteorological parameters was investigated. The result of the correlation between temperature and rainfall analyzed for summer is R^2>0.62 and R^2>0.56. No significant relationship was found for precipitation in winter while it is significant for temperature. In the study, based on the curves of graphics drawn using the *Clausius-Clapeyron* equation, the relationship between gas phase concentrations of atmospheric i-PCBs and air temperature was found to be significant. In addition, according to *Clausius-Clapeyron* curves, gas phase concentration of i-PCBs, there is a significant relation in especially summertime. Higher i-PCB concentrations are also related to the velocity and direction of the prevalent wind. According to wind direction, PCB concentrations can increase or decrease.

Then, according to *Clausius-Clapeyron* curves, the ambient temperature and evaporation from the contaminated areas together with the temperature are thought to contribute to the measured PCB concentrations. It was also observed when the effect of meteorological factors on atmospheric PCB components was taken into consideration that temperature contributed significantly to the measured PCB concentrations. It was observed that high concentration values of PCBs are generally related to the prevalent wind and speed. In addition, it was determined that PCB rates increase or decrease according to the wind directions. It was also observed that precipitation in summer and winter periods affected PCB concentrations. It is seen that atmospheric PCB concentrations decrease especially in the months when the rainfall is high and vice versa during the summer months when the rainfall is low.

As a result, all these meteorological factors, especially temperature, were determined to affect i-PCB concentrations significantly. This result was attributed to the variability of the meteorological conditions during sampling and the dispersed structures of possible PCB resources. The results of the study were found to be convenient with those given in literature in terms of the distribution of dominant i-PCB component and homologous group and dominant phase distribution. In addition, higher concentrations of i-PCBs were not observed in Bayburt compared to other studies. Even though the concentrations of i-PCBs were calculated to be lower in Bayburt, it is thought to be important to monitor and control i-PCBs in urban environments since they are evaluated to be toxic and carcinogenic.

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