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Contents

ARKTİK SALINIMIN TÜRKİYE'DEKİ İSTASYONLARIN AYLIK MİNİMUM ORTALAMA SICAKLIK DEĞERLERİ ÜZERİNDEKİ ETKİLERİNİN AYLIK ANALİZİ 1

DETERMINATION OF AEROSOLS AND CLOUDS FROM ALL-SKY IMAGES FOR THE ESTIMATION AND FORECASTING OF SOLAR IRRADIANCE 11

WAVELET METHODS FOR INVERSE PROBLEM IN ATMOSPHERİC SCİENCES 12

HALE OLUŞUMUNUN HALOSIM3.6 HALE SİMÜLASYON PROGRAMIYLA İNCELENMESİ 14

BU DÜNYA HEPİMİZİN 20

YEAR-ROUND VARIABILITY IN THE MIDDLE ATMOSPHERIC TRANSPORT 24

Isınma ve Soğuma Derece Günlerin Elektrik Tüketimi Üzerindeki Etkisi 29

ATMOSFERİK TAHMİNLERİN DENİZ ORTAMINDA DOĞRULUK ANALİZİ 40

BIDIMENSIONAL DIAGNOSTICS OF ATMOSPHERIC BLOCKING AND IMPACT OF BLOCKING ON REGIONAL WEATHER 50

HIGH IMPACT WEATHER: THE ANTALYA 9 OCTOBER 2011 FLASH-FLOODING EVENT 60

SPATIOTEMPORAL VARIATION OF EXTREME RAINFALL EVENTS OVER HIMACHAL PRADESH 72

TÜRKİYE'DE HORTUM VAKALARININ ZAMANSAL VE MEKÂNSAL ANALİZİ 79

Elektrik ve Manyetik Alan Sinyalleri Kullanarak Yıldırım ve Şimşek Olaylarının Belirlenmesi 86

ŞEHİR SELLERİ TAHMİN VE ERKEN UYARI MODELİ 95

İSTANBUL ANA ULAŞIM YOLLARI BUZLANMA RİSK HARİTALARI 105

INTEGRATED MODELING OF AIR POLLUTION AND ASSOCIATED HEALTH IMPACTS 113

Challenges in Fighting Air Pollution 114

Aerosols in the Mediterranean Region and Middle East: Patterns and impacts 115

ERZURUM KENT MERKEZİNDE ISINMA KAYNAKLI PAH'LARIN ATMOSFERİK KONSANTRASYONLARI. 116

POLİKLORLU BİFENİLLER (PCB'LER)'İN ERZURUM KENT ATMOSFERİNDE GÖZLENEN KONSANTRASYONLARI 124

METEOROLOJİK KOŞULLARIN HAVA KİRLİLİĞİ ÜZERİNE ETKİLERİNİN İNCELENMESİ: ANKARA İLİ ÖRNEĞİ 132

SYNTHESIS OF A FUEL BIOADDITIVE BY THE CATALYTIC MEMBRANE FOR IMPROVEMENT OF COMBUSTION QUALITY 139

HAVA KALİTESİ ve DEĞERLENDİRMESİNDE MODELLEME YAKLAŞIMI 147

DIURNAL VARIATION OF ORGANIC AEROSOL IN URBAN EAST ST. LOUIS, IL 157

AN INVESTIGATION OF THE RELATIONSHIP BETWEEN MODIS AOD AND $PM_{2.5}$ OVER MARMARA REGION, TURKEY 170

Determination of mercury in Istanbul airborne particles using solid sampling HR-CS GFAAS 181

A novel approach for the determination of fluorine and sulphur in airborne particles by HR-CSAAS 189

MEASURED VOC CONCENTRATIONS IN BALIKESIR AMBIENT ATMOSPHERE 198

ANKARA BÖLGESİ HAVA KİRLİLİĞİ ANALİZİ VE MODELLEMESİ 204

ESKİŞEHİR' de KENTSEL ve YARI-KENTSEL İKİ NOKTADA ATMOSFERİK ORGANİK KİRLETİCİLERİN ÖLÇÜMÜ 212
KÜTAHYA KENT VE KIRSALINDA ÖLÇÜLEN NO_x, SO_2 ve O_3 DERİŞİMLERİNİN DEĞERLENDİRİLMESİ 221
BALIKESİR'DE TAŞINIM METEOROLOJİK ETKİLER KAPSAMINDA HAVA KİRLİLİĞİ ARAŞTIRMASI 231
THE INFLUENCE OF RELATIVE HUMIDITY ON CONCENTRATIONS ($PM_{10,}$ TSP) IN BAGHDAD CITY 239
Ankara ve Çevre İllerde Hava Kirliliği Dağılımı ve Emisyon Envanterinin Belirlenmesi 251
IMPACT OF MARINE AEROSOLS IN FRONTAL ZONES OVER OCEAN SURFACE 261
MODIS AEROSOL ÜRÜNLERİ KULANILARAK PM10 KONSANTRASYONU TAHMİNİ 271
SEASONAL VARIATION OF GAS/PARTICLE PARTITIONING OF PCB COMPOUNDS IN THE ISTANBUL ATMOSPHERE 281
Comparison of Different Mixing Height Estimation Models for Air Pollution Modeling Studies 291
Chemical Composition of Precipitation in Three Different Parts of Turkey 300
Spatial Variation of PM10 in Turkey 311
HAVA KALİTESİ VE METEOROLOJİK MODELLERİN ONLİNE ENTEGRASYONU: TÜRKİYE ÖRNEĞİ 324
CBS YARDIMIYLA EMİSYON ENVANTERİ OLUŞTURULMASI: ANKARA ÖRNEĞİ 335
Investigation into relationships among NO, NO_2, and O_3, at a coastal rural/suburban site in Istanbul 348
Atmospheric Gravity Waves in Aviation 359
Diurnal valley winds and exchange processes over mountainous terrain 380
Birleşik Arab Emirlikleri'ndeki Toz Fırtınası, 28 Şubat 2009 381
YANARDAĞ PATLAMALARININ TÜRKİYE'DEKİ FIR SAHALARI ÜZERİNE ETKİSİ 393
HAVACILIK İÇİN ÖNEMLİ ATMOSFERİK SEVİYELER 405
METEOROLOJİK AÇIDAN HAVALİMANLARIMIZIN STATÜLERİ, KATEGORİLERİ ve GÖZLEM SİSTEMLERİ 413
CODA RAPORLARININ İNCELENMESİ 423
AVRUPA BÖLGESİ İÇİN KAZA-KIRIM RAPORLARININ İNCELENMESİ 434
SAYISAL HAVA TAHMİN MODELLERİ KULLANILARAK BUZLANMA ANALİZ VE TAHMİNİ 442
İZMİR ADNAN MENDERES HAVALİMANI'NIN KÜMÜLONİMBÜS (Cb), ORAJ VE SİS ANALİZİ 452
15 TEMMUZ 2013 TARİHİNDE ANKARA ESENBOĞA HAVALİMANI'NDA YAŞANAN BORAN OLAYININ İNCELENMESİ 465
TAF Yayınlarında TEMPO Periyodları ile Tahmin Edilen Meteorolojik Değişkenlerin Üç Meydan İçin Tutarlılık Analizi 475
SON 10 YILDA AVRUPAYI VE ÜLKEMİZİ ETKİLEYEN VOLKANİK OLAYLARIN HAVACILIK METEOROLOJİSİ BAKIMINDAN İNCELENMESİ 487
TOPRAK VE RÜZGAR ESASLI ISITMA-SOĞUTMA SİSTEMLERİNİN LABORATUVAR ÖLÇEKLİ İNCELENMESİ 498
METEOROLOJİK OLAYLAR İLE DEPREM İLİŞKİSİ; İstanbul Örneği 502

HAVACILIK SEKTÖRÜNDE İNSAN KAYNAKLARI AÇISINDAN BAKIM ORGANİZASYONLARINDA ÇALIŞANLARIN EĞİTİMİ 515
COĞRAFİ BİLGİ SİSTEMLERİ YARDIMIYLA HAVZA KARAKTERİSTİKLERİNİN BELİRLENMESİ 521
SAMSUN – KÜRTÜN ÇAYI TAŞKIN HESABI 529
Yamula Barajı'nın Havacılık İçin Önem Arz Eden Meteorolojik HadiselerÜzerindeki Etkisi 537
KONYA SELÇUKLU TEPEKENT GÖLETİ TAŞKIN HİDROLOJİSİ 548
ANİ TAŞKIN ERKEN UYARI SİSTEMİ 559
PROJE KESİTLERİNDE AKIM ÖLÇÜMLERİNİN ÖNEMİ 570
VULNERABILITY OF AN AQUIFER IN AN ARID ENVIRONMENT 577
KAHRAMANMARAŞ MERKEZ ÇAMLIBEL GÖLETİ SU POTANSİYELİ HESABI VE SULAMA ALANININ BELİRLENMESİ 589
BEHAVIOUR OF POLLUTANTS IN URBAN STORM WATER SANITATION NETWORKS (FEATURES AND ORIGINS): CASE THE PLAIN OF ANNABA 597
ORTALAMA SICAKLIKLARDAKİ TÜRDEŞLİK KIRILMALARININ İKLİMGÖSTERGELERİYLE İLİŞKİSİ..... 605
İSTANBUL'DA HAKİM RÜZGARIN PERSİSTANSINDAKİ DEĞİŞİMLERİN YÖNSEL İSTATİSTİK YÖNTEMİ İLE İNCELENMESİ..... 615
TÜRKİYE YAĞIŞLARININ SPEKTRAL-KÜMELEME YÖNTEMİ İLE SINIFLANDIRILMASI 627
MPI-ESM-MR MODELİNİN RCP4.5 VE RCP8.5 SENARYOLARINA GÖRE SICAKLIK ve YAĞIŞ PROJEKSİYONLARI 636
HATAY'DA YAĞIŞLARIN TREND ANALİZİ TREND ANALYSIS OF PRECIPITATION IN HATAY..... 646
KÜRESEL ISINMANIN GÜNEŞ ENERJİSİNE ETKİLERİNİN LABORATUVAR ÖLÇEKLİ İNCELENMESİ..... 657
URBANIZATION EFFECT ON TRENDS OF EXTREME TEMPERATURE INDICES IN ANKARA 664
İKLİM DEĞİŞİKLİĞİ VE BUNA BAĞLI AFETLER YOL HARİTASI BELGESİ..... 676
Greenhouse Gas Mitigation Works and Measures Taken Against Climate Change: Case of Istanbul 686
HAVACILIK AMAÇLI METEOROLOJİK MESAJLAR..... 698
KARADENİZ'DE ASİDİFİKASYONUN UZUN DÖNEMLİ ANALİZİ VE TÜRKİYE İKLİM DEĞİŞİMLERİ İLE İLİŞKİLENDİRİLMESİ 706
URBAN CLIMATE IMPACT ANALYSIS USING WRF-CHEM AND DIAGNOSTIC DOWNSCALING TECHNIQUES..... 716
TÜRKİYE'DE İKLİM DEĞİŞİKLİKLERİNİN PEYZAJ VE KENTSEL PLANLAMA ÇALIŞMALARINA OLASI ETKİLERİ VE OLUMSUZLUKLARA KARŞI ALINABİLECEK ÖNLEMLER..... 726
ANALYSIS OF WIND SPEED VARIABILITY IN TURKEY VIA MANN-KENDALL STATISTICS..... 732
TRENDS IN DAILY CLIMATE EXTREMES IN MARMARA REGION (NW TURKEY) AND THEIR CONNECTION TO THE ATMOSPHERIC CIRCULATION TYPES 744
KENTSEL İKLİM DEĞİŞİKLİĞİ LİTERATÜRÜ; DÜNYA VE TÜRKİYE'DEN SEÇMELER 759
KÜRESEL İKLİM DEĞİŞİMİNİN D.KARADENİZ İLLERİNDE HİSSEDİLMESİ ÖRNEĞİ..... 771
ERAfrica Project: LOCLIM3- The local climate changes in 3 cities of Cairo, Nairobi and İstanbul: İstanbul Case 781

TÜRKİYE GENELİ YAĞIŞ VE SICAKLIK VERİLERİNDE EĞİLİMLER VE SALINIMLAR 791
TÜRKİYE'DEKİ OLASI İKLİM DEĞİŞİKLİĞİ ETKİLERİNİN MANN-KENDALL TREND YAKLAŞIMI İLE İNCELENMESİ 801
İKLİM DEĞİŞİKLİĞİNİN ANLIK YÜKSEK AKIMLARA ETKİSİ 812
İRAN'DA ENTERPOLE EDİLMİŞ ERA-40 REANALİZ VERİ SETİNE AİT AYLIK ORTALAMA SICAKLIK VE AYLIK TOPLAM YAĞIŞIN DEĞERLENDİRİLMESİ 823
TÜRKİYE'DE ULUSAL VE BÖLGESEL KALKINMA PLANLARINDA İKLİM DEĞİŞİKLİĞİNE DAİR DUYARLILIKLAR 832
Evaluation of Mesoscale Events using dual-Polarization and Doppler Radar Observations 840
AEROSOL OPTİK DERİNLİĞİ VERİLERİNİN TÜRKİYE İÇİN ALANSAL VE ZAMANSAL DEĞİŞİMLERİNİN İNCELENMESİ 841
METEOSAT İKİNCİ NESİL UYDULARI (MSG) YANGIN ÜRÜNÜNÜN BÖLGESEL EŞIK DEĞERLERI İLE TÜRKIYE COĞRAFYASI İÇIN İYİLEŞTİRİLMESİ 853
ESTIMATION OF THE PRECIPITABLE WATER VAPOR FROM GNSS OBSERVABLES 863
Meteoroloji Radarlarında Kullanılan Farklı Yağış Hesaplama Yöntemlerinin Çeşitli Yağış Olayları İçin Karşılaştırılması 874
SAMSUN METEOROLOJİ RADARININ HAVZA SU YÖNETİMİNDE KULLANIMI: 3 TEMMUZ 2012 ÖRNEK OLAY İNCELEMESİ 885
EFFECTS OF THE ETESIAN WIND REGIME ON COASTAL UPWELLING, FLOODS AND FOREST FIRES IN THE SEAS OF THE OLD WORLD 895
19/07/2013 HATAY DÖRTYOL'DAKİ SEL AFETİ 900
STANDARDİZE YAĞIŞ – EVAPOTRANSPİRASYON İNDİSİ (SPEI) İLE KONYA HAVZASI KURAKLIK ANALİZİ 907
BATI AKDENİZ BÖLGESİNDE METEOROLOJİK AFET RİSKLERİNDEN KORUNMAK İÇİNİKLİM PARAMETRELERİNE DAYALI SERA TASARIMI 918
FARKLI ZAMAN ÖLÇEKLERİNDEKİ STANDART YAĞIŞİNDEKSLERİNİN HAVZA BAZINDA DEĞERLENDİRİLMESİ 930
ÇERNOBİL FELAKETİNİN METEOROLOJİK VERİLERLE DEĞERLENDİRİLMESİ 941
LOWESS YÖNTEMİNİN ATMOSFERİK SÜREÇLERE UYGULANMASI: BUHARLAŞMA-ATMOSFER BASINCI İLİŞKİSİ 952
ZİLAN HAVZASI (VAN, TÜRKİYE) HİDRO-METEOROLOJİK ÖZELLİKLERİNİN İNCELENMESİ 960
DELİÇAY VE BENDİMAHİ HAVZALARINDA (VAN, TÜRKİYE) AKIM TRENDLERİNİN DEĞERLENDİRİLMESİ 970
İstanbul-Kandilli İkliminin 1912-2014 Yılları Arasında Salınım ve Eğilimleri 978
ANKARA'DA FARKLI YÖNTEMLERLE YAPILAN OZON PROFİL ÖLÇÜMLERİNİN KARŞILAŞTIRILMASI 988
A NEW MESOSCALE ENSEMBLE PREDICTION SYSTEM: SCIENTIFIC DESCRIPTION AND PRELIMINARY RESULTS 1000
INVESTIGATION OF THE IMPACT OF LAND USE MODIFICATION TO THE URBAN HEAT ISLAND PHENOMENA USING WRF OVER ISTANBUL 1011

WRF ve ALARO SAYISAL HAVA TAHMİN MODELLERİ İÇİN VERİFİKASYON SONUÇLARININ KARŞILAŞTIRILMASI 1021
Kuvvetli yağışlarda GPS-IPWV (Yağışabilir su miktarı) verisinin önemi ve kullanılması 1030
A CASE STUDY OF ONLINE ENVIRO-HIRLAM AND WRF-CHEM MODEL DURING AN AIR POLLUTION EPISODE IN ISTANBUL 1039
Analysis of Rainfall Hazard in the City of Algeria 1049
BİYOKLİMATİK KONFOR; TARİHİ GELİŞİMİ ve TÜRKİYE ÖRNEKLERİ 1057
THE DEGRADATION OF POLLUTANTS IN DIFFERENT RECEIVING ENVIRONMENTS: CASE OF THE PLAIN OF ANNABA 1067
AKDENİZ BÖLGESİ ADANA, ANTALYA, MERSİN VE ISPARTA İSTASYONLARI MİNİMUM SICAKLIK EĞİLİMLERİ 1077
2013–2014 TARIM YILI KURAKLIK ANALİZİ 1086
TÜRKİYE'DE TOPRAK SICAKLIĞI YÖNÜNDEN SERİN İKLİM TAHILLARIN EKİM ZAMANININ BELİRLENMESİ 1098
VEJETATİF ORTAMDA TOPRAK ISI AKISI, TERMAL İLETKENLİK VE TERMAL YAYILIMIN ZAMANSAL DEĞİŞİMİNİN ANALİZİ 1106
KARLA KAPLI BİR YÜZEYİN ENERJİ DENGESİNİN İNCELENMESİ 1114
DENİZLİ İLİNDE İKLİM İSTEKLERİNE GÖRE FARKLI ÜZÜM ÇESİTLERİNİN YETİŞTİRİLEBİLECEĞİ ALANLARIN BELİRLENMESİ 1124
BİTKİ SOĞUKLAMA İSTEĞİ HESAPLAMA PROGRAMI (BİSİP) 1138
AYÇİÇEĞİ BİTKİSİNİN KARBON DEĞİŞİMİ İLE TOPRAK SU İÇERİĞİ ARASINDAKİ ETKİLEŞİMİN ANALİZİ 1146
SICAKLIK-NEM İNDEKSİ PROGRAMI (SINEP) 1156
Eddy Kovaryans Yöntemi ile Karpuz Bitkisinin CO_2 Akısının Belirlenmesi 1166
Downscaling AMSR_E-Derived Soil Moisture Using MODIS Data 1173
BUĞDAY BİTKİSİNİN HASAT ÖNCESİ VE SONRASI KARBON AKILARININ DEĞİŞİMİNİN İNCELENMESİ 1182
AQUACROP MODELİ İLE AYÇİÇEĞİ BİTKİSİ GELİŞİMİNİN İKİ FARKLI LOKASYONDAKİ SİMÜLASYONU 1189
Evapotranspirasyon'un Toprak ve Su Faktörlerindeki Değişimlere Hassasiyetinin Model İle Belirlenmesi 1196
On the use of lidars in wind energy 1204
MUĞLA İÇİN ATMOSFER GEÇİRGENLİĞİNDEN YARARLANILARAK SAATLİK DİREKT RADYASYON TAHMİNİ 1205
Rüzgar Türbinlerinde Türbülans Yoğunluğu ve 1.5 kW'lık Sistem için Uygulama 1214
WATER HYACINTH: A SUSTAINABLE BIOMASS TO ENERGY AN INDIAN PERSPECTIVE 1221
İstanbul İçin Eğimli Yüzeye Gelen Günlük Toplam Radyasyon Tahmini 1232
TÜRKİYE GENELİNDE RÜZGAR HIZI PERSİSTANSININ FARKLI YÖNTEMLERLE İNCELENMESİ 1244
ENERJİ, ENTROPİ, HAVA VE İKLİM 1253
NOKTASAL TOPLAM YARI VARYOGRAM (NTYV) YÖNTEMİ İLE RÜZGAR ALAN MODELLEMESİ 1263

Deniz Etkisi Kar Yağışları ve Gök Gürültüsü 7 Ocak 2015 Trabzon Örneği 1271

SEASONAL VARIATION OF GAS/PARTICLE PARTITIONING OF PCB COMPOUNDS IN THE ISTANBUL ATMOSPHERE

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ÖZET

Bu çalışmada, dioksin benzeri ve indikatör PCB'lerin gaz/partikül dağılımının mevsimsel değişimi araştırılmıştır. Gaz ve partikül faz örnekleri yüksek hacimli hava örnekleyici ile Mayıs 2011 ve Ekim 2013 tarihleri arasında toplanmıştır. DB-PCB'ler gaz fazında bulunma eğilimi (%77) göstermiş olup yazın gaz fazı oranında artış gözlenmiştir. Gaz fazı oranları yaz mevsimi için %87, kış mevsimi için %70 olarak belirlenmiştir. Db-PCB'lere benzer şekilde, indikatör PCB'lerde gaz fazında (%90) bulunma eğilimi göstermiştir. Db-PCB'lerin aksine indikatör PCB'lerin gaz/partikül dağılımı mevsimsel olarak önemli bir değişim göstermemiştir. Kış, yaz, sonbahar ve ilkbahar için gaz fazının toplam konsantrasyondaki oranı sırasıyla %88, %89, %87 ve %84 olarak belirlenmiştir. Partikül fazı oranı ise yazın %12'si, kışın ise %16 olarak belirlenmiştir. Db-PCB'ler için sıcaklık, yağış ve rüzgar hızı, SR ve UV arasında negatif yönlü zayıf korelasyon basınç için ise pozitif yönlü zayıf korelasyon belirlenmiştir. İndikatör PCB'ler için yağış ile pozitif yönlü korelasyon, rüzgar hızı ve basınç için negatif yönlü zayıf korelasyon belirlenmiştir.

Anahtar Kelimeler: Poliklorlubifenil, gaz/partikül dağılımı, atmosferik örnekleme

ABSTRACT

In this study, gas/particle partitioning of dioxin-like PCBs (Dl-PCB) and indicator PCB was investigated at three sampling areas of Istanbul. Gas and particle phase samples were collected via high volume air sampler on monthly time intervals from May 2011 to October 2013. Dl-PCB compounds showed a tendency of existing in gas phase (77%) with increasing trend in the summer season. The percentages of gas phase were determined to be 88% and 66% for summer and winter seasons, respectively. Indicator PCBs showed a tendency of existing in gas phase (87%) similar to Db-PCB compounds. Unlike Dl-PCBs, gas/particle partitioning of indicator PCBs did not show significant seasonal variation. Gas phase ratios of indicator PCBs were detected to be 88%, 89%, 87%, 84% for winter, summer, autumn and spring seasons, respectively. Low negative correlations were found between temperature, precipitation, wind rate, solar radiation (SR), ultra violet (UV) and Dl-PCB concentrations. Similarly, low negative correlations were found between wind rate, atmospheric pressure and indicator PCB compounds.

Key Words: Polychlorinated biphenyl, gas/particle partitioning, atmospheric sampling

INTRODUCTION

Polychlorinated biphenyls (PCBs) are a group of organic chemicals which can be odorless or mildly aromatic solids or oily liquids. The 12 PCBs behave through the aryl hydrocarbon receptor (AHR) to cause the full range of toxic responses revealed by 2,3,7,8-tetrachlorodibenzo-p-dioxin. Hence, these PCB congeners are referred to as the dioxin-like PCBs. The 197 PCB congeners which except for dioxin-like PCB are currently referred to collectively as indicator congeners. Examples of primary PCB sources include landfills, hazardous waste sites, incineration of PCB-containing wastes, leakage from old electrical equipment (capacitor, transformer etc.) and improper disposal or spills (ATSDR, 2000). Leaks from capacitors have been identified as the most important

source of PCBs in the UK (Dyke, 1997). Other PCB sources include the various combustion processes such as steel production, coal combustion, sinter production, application of sewage sludge to land, waste incineration etc. In addition to major source of PCB release to the environment involves the volatilization from surface waters, soil and sediment into the atmosphere and subsequent removal from the atmosphere via wet/dry deposition and then revolatilization. Therefore it has been reported that urban and industrial areas are the main sources of the PCB compounds (Brunciak et al., 2001; Ozcan and Aydin, 2009). PCBs will primarily exist in the gas-phase after released to the atmosphere; however tend to have a particle phase with increasing chlorination degrees. Gas/particle partitioning of semi volatile organic compoundsaffects the atmospheric transportation, atmospheric residence time and atmospheric removal processes of theirs (Matsumato et al., 2010). In addition to gas/particle partitioning of PCB compounds is influenced by many factors such as ambient air temperature, vapor pressure of organic compounds, total suspended solid concentration and properties of the particle matters (size distribution, elemental or organic carbon content).

In this study, gas/particle partitioning and seasonal variations of dioxin-like PCB (dl-PCB) and indicator PCB concentrations were investigated at three different areas in Istanbul. In addition, relationship between atmospheric concentration levels and meteorological parameters (temperature, solar radiation, solar energy, UV, precipitation) were also evaluated by using statistical analysis.

MATERIAL AND METHOD

SITE DESCRIPTION

Ambient air samples were collected at three different sampling stations in Istanbul which is located on the north-western part of Turkey (see Fig. 1). The city has about 14 million population with various anthropogenic activities in the sense of PCDD/F sources such as dense motor vehicle traffic and mixed groups of industrial facilities. Therefore, three different sampling locations were chosen in this study. Davutpasa sampling region (latitude: 41.02 N; longitude: 28.82 E) involves residential and industrial structures together with Turkey's biggest intercity bus terminal and foundry industrial zone which are located about 1.5 km NE and 9 km NW of the sampling point, respectively. Yıldız sampling station (latitude: 41.30 N; longitude: 29.00 E) is located in the city center which receives motor vehicle emissions due to dense public transportation and ship emissions from the Bosphorus marine traffic. Fenertepe sampling station (latitude: 41.90 N; longitude: 28.47 E) is located out of the city center and surrounded by large forest area. Although there is no industrial activity in the vicinity of this location, there are medical and hazardous waste incineration plants which are located about 8 km NE and 12 km SE of this sampling station, respectively.

Fig. 1. Location of sampling stations.

SAMPLING AND ANALYSIS

Sampling and analysis were conducted in accordance with the reference method of EPA TO-9A. Gas and particle phase samples were collected simultaneously at three sampling points during May 2011 to October 2013 using high volume sampler. High volume air samplers were operated at 0.225 m^3 min^{-1} flow rate. They were operated for a period of 5–7 d (120–168 h) at each sampling time. High volume samplers were placed at heights of about 10 m, 3 m and 15 m above ground level for Davutpaşa, Yıldız and Fenertepe sampling stations, respectively. Quartz fiber (QF) filters (10 cm dia) and polyurethane foam (PUF) cylindrical filters (6.0 cm-dia) were used for particle and gas phase samplings, respectively. Before sampling, QF filters were conditioned at 450°C for 5 h and PUF filters were pre-cleaned with acetone in soxhletextractor for 16 h. Extraction, clean up and fractionation were applied for QF and PUF filters after sampling. PUF and QF filter were extracted separately with toluene for 20 h using soxhlet extractor. After extraction, toluene extract is concentrated in rotary vacuum evaporator, re-dissolved in hexane and pre-cleaned by shaking with 10 mL concentrated sulfuric acid at laboratory temperature. Pre-cleaned hexane extract was transferred on top of multi-layer silica gel column and eluted with hexane. Extract was concentrated with modified Kuderna–Danish concentrator up to 0.5–1 mL. For fractionation was used 1g florisil column and combination of hexane and dichloromethane for elution of PCDD/F and PCB fractions. First hexane fraction containing mono-ortho PCBs and non dioxin-like PCBs is concentrated using Kuderna-Danish concentrator to volume about 0,5 ml. Final extractis concentrated up to dryness under nitrogen stream and then 2-to-4 µL of samples were injected to high-resolution gas chromatography/high-resolution mass spectrometry (HRGC–HRMS, Thermo Gas Chromatograph Trace GC Ultra-HRMS) for the chromatographic analysis ofdl-PCB and indicator PCB compounds.$^{13}C_{12}$-labeled sampling andextractionstandards were used to determine the recovery efficiency of sampling and extraction processes. Recovery values were determined in the rangesof 71–94% and 64–114% for sampling and extraction standards, respectively.

RESULTS AND DISCUSSIONS

CONCENTRATION LEVELS OF DB-PCB$_S$ AND NON- DB-PCB$_S$ IN THE AMBIENT AIR

Average concentrations of dl-PCB compounds were detected to be 4.81 (3.1-8.3) pg m^{-3} according to the results of measurements conducted between May 2011 and October 2013. When the change of concentration was evaluated according to the sampling stations, the average concentrations of these compounds were found to be 6.13 (3.35-9.8) pg m^{-3}, 5.06 (2.5-8.02) pg $m^{-3}$and 3.24 (0.34-7.3) pg m^{-3} for Davutpaşa, Yıldız and Fenertepe sampling stations, respectively. Average I-TEQ concentrations were calculated to be 10.7 fg m^{-3}, 7 fg m^{-3} and 3.75 fg m^{-3} for Davutpaşa, Yıldız and Fenertepe sampling stations, respectively.The concentration values of indicator PCBs ranged from 19 pg m^{-3} to 170 pg m^{-3} that the average concentration was detected to be 42 pg m^{-3} according to the results of all samplings. Average concentrations were detected to be 60 pg m^{-3} (24-407 pg m^{-3}), 38 pg m^{-3} (12-85 pg m^{-3}) and 30 pg m^{-3} (15-81 pg m^{-3}) at Davutpaşa, Yıldız and Fenertepe sampling stations, respectively. Indicator PCB_s concentration was increased at Davutpaşa sampling station at April 2012 sampling period. The average concentration was found to be 406 pg m^{-3} at this period. However, significant increase was not observed for dl-PCB_s concentration. The concentration was detected to be 75 pg $m^{-3}$and 29 pg m^{-3} at Yıldız and Fenertepe sampling stations during the April 2012 sampling period, respectively. The observed fire in the textile factory where is the located close to Davutpaşa sampling station was considered to be the cause of the high concentration level during this sampling period. As it is known, PCBs are formed by the various processes including industrial production processes, incineration of the PCB-containing materials and other thermal processes at high temperature (Aries et al., 2004; Choi et al., 2008).

GAS/PARTICLE PARTITIONING OF DB-PCB$_S$ AND NON DB-PCB$_S$ COMPOUNDS

Dl-PCBs compounds showed a tendency to be existed in gas phase at high percentage (77%). Gas and particle phase concentrations for dl-PCB compounds were measured to be 5130 fg m^{-3} and 1521 fg m^{-3}, respectively. PCB-118 (%41), PCB-105 (15%) were detected to be the most abundant congeners in a gas phase while PCB-118 (29%), PCB-189 (15%), PCB-105 (13%) were detected commonly in particle phase. The ratio of gas phase concentrations to total average concentrations were calculated to be 69%, 80%, 82%, whereas the particle phase ratios were calculated to be 31%, 20%, 18% for Davutpaşa, Yıldız and Fenertepe sampling stations, respectively. Gas/particle partitioning of PCDD/F compounds were investigated in the same sudy that they were found in a particle phase (92%) unlike Dl-PCBs compounds. This situation can be explained by the differences in a their formation mechanisms, formation temperature and physicochemical properties such as vapor pressure, octanol water coefficient and octanol air coefficient etc.

Similar to the dl-PCBs, indicator PCBs showed a tendency to be existed in gas phase (90%). Average concentrations of indicator PCBs were calculated to be 90 pg m^{-3} for gas phase and 10 pg m^{-3} for particle phase according to the all sampling results. The average percentages of gas phase concentrations were found to be 92%, 88% and 90% for Davutpaşa, Yıldız and Fenertepe sampling stations, respectively. The same congeners (PCB 28, PCB 52, PCB 101) were found to be dominant for both gas phase and particle phase. Highest gas phase ratios were detected for PCB-28 (43%), PCB-52 (21%), PCB-101 (11%) congeners whereas the highest particle phase ratios were detected for PCB-28 (26%), PCB-52 (13%), PCB-101 (12%) congeners.

EFFECTS OF THE SEASONAL VARIATION ON GAS/PARTICLE PARTITIONING

Seasonal variations of dl-PCB_s and indicator PCB_s were shown in Fig 2. Significant seasonal variation was not observed for dl-PCB compounds. Highest average concentrations were observed in winter and autumn seasons. There was no significant difference between the concentrataion levels of indicator PCB_s for autumn, summer and winter seasons while the concentration value for spring season was higher than the other seasons (Fig.2). The reason of this situation may be related to the textile factory fire during the April 2012 sampling period as described in the previous section. As a result, dl-PCB_s and indicator PCB_s concentrations did not show significant seasonal variations. In particular, the fluctuations in concentrations of dioxin-like PCB_s were much smaller than those of indicator PCB_s. It was reported that the concentration of PCB_s compounds in summer is higher than winter (Fiedler, 1999; Castro-Jimenez, 2009) unlike this study.

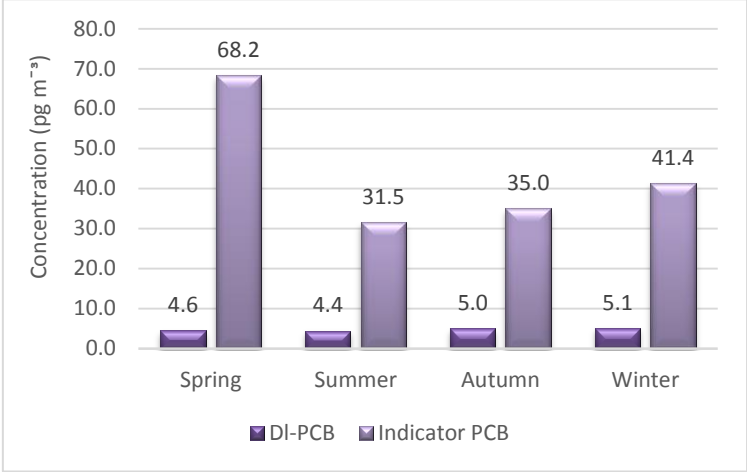

Fig.2 Sesonal variation of PCB compounds

Seasonal variation of gas/particle partitioning of dl-PCB_s was shown in Fig.3. Average gas phase concentrations of dl-PCBs were detected to be 3872 fg m^3 and 3577 fg m^{-3} whereas particle phase concentrations were detected to be 1566 fg m^{-3} and 557 fg m^{-3} for winter and summer seasons, respectively. In addition to the gas phase ratio of dl-PCB compounds increased in summer season. Gas phase ratios were detected to be 87%, 70%, 79%, 84% for summer, winter, autumn and spring seasons, respectively. Particle phase ratio of dl-PCB compounds was detected higher in winter season (30%) than summer season (13%) unlike gas phase ratio. Kim et al. (2011) reported that gas phase ratio of dl-PCB compounds was found to be 97-99% and 23-62% for summer and winter seasons.The observed increase in summer season can be explained by the desorption of PCB compounds from the reservoir sources such as soil, water, vegetation (Halsall et al., 1995; Manchester-Neesving and Andren, 1989; Hillery et al., 1997; Simcik et al., 1997, 1999; Cousins and Jones, 1998; Wania et al., 1998). PCB compounds would be desorbed more easily from the soil in summer season and they can easily be transported over long distances at high temperatures on a global scale (Taşdemir et al., 1997). Additionally, these compounds can be removed from the atmosphere by snowfall and rainfall during especially in winter season (Daly and Wania, 2004). Particle phase concentration of dl-PCBs increased in winter season. This situation can be explained by (1) adsorption of the gas phase dl-PCBs on to the particle surface due to the low ambient air temperature in winter season (2) low mixing height in winter season. PCBs in the particle phase are removed by particle washout mechanism. The dissolution of gaseous PCBs into rain and cloud droplets depend on the

Henry's Law constant and air/water partition coefficient calculated from the ratio of vapour pressure to water solubility (Bidleman, 1988).

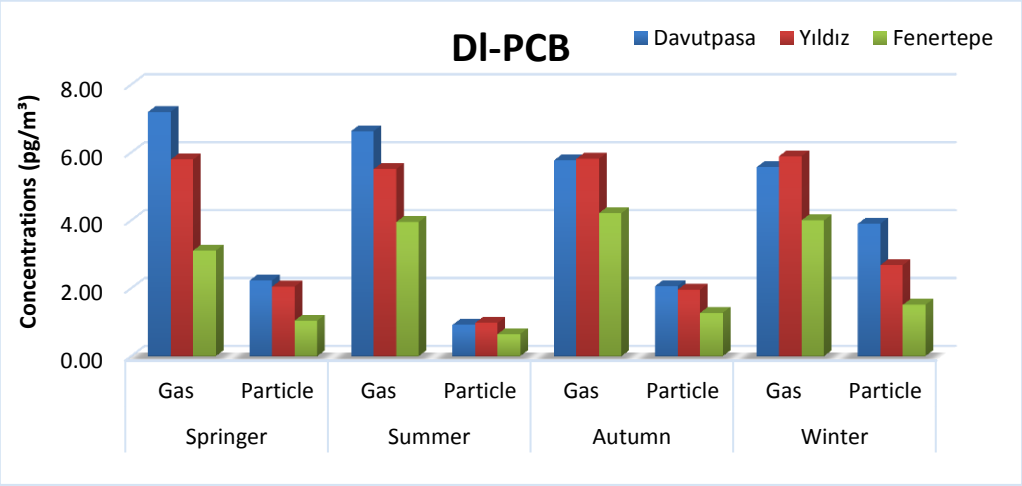

Fig.3 Seasonal variation of gas/particle partitioning of Dl-PCB$_s$

PCB 118 (60%), PCB 105 (21%), PCB 77 (9%) and PCB 156 (5%) were detected to be the most abundant congeners for gas phase during the summer season (Fig.4). The similar congeners were determined to be dominant also for particle phase. The ratios of PCB 118, PCB 105, PCB 77 and PCB 156 congeners were found to be 55%, 24%, 11% and 6% for particle phase in summer season. Similar to the summer season, PCB 118 (38%), PCB 105 (21%), PCB 156 (10%), PCB 77 (9%) were detected as primary congeners in winter. PCB 118 was reported as predominant congener by several researchers (Gatehouse, 2004; Kim and Masunaga, 2004). PCB 118 is a dominant congener in Kanechlor products, which were first produced in 1954 by Kaneka Corporation in Japan as commercial PCB products (Ikenaka et al., 2003).

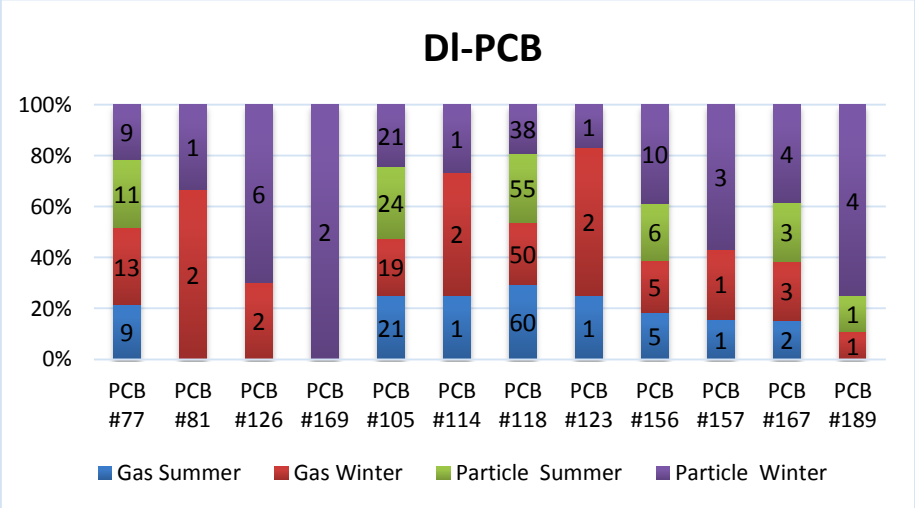

Fig.4 Gas/particle distribution of dl-PCB$_s$ congeners

Gas/particle partitioning of the indicator PCBs was shown in Fig.5. Average gas phase concentrations were measured to be 28 pg m^{-3} and 36 pg m^{-3} while particle phase concentrations were measured to be 3.3 pg m^{-3} and

5 pg m^{-3} for summer and winter seasons, respectively. Several previous publications was reported that the gas phase concentration of PCBs congeners are higher than particle phase concentration (Mandalakis et al., 2002; Taşdemir et al., 2004; Cindoruk et al., 2007). The ratio of gas phase concentration to total gas phase concentration were found to be 88%, 89%, 87% and 84% in winter, summer, autumn and spring seasons, respectively. Gas phase ratios of PCB 28, PCB 52, PCB 101 congeners were higher than the other congeners in summer and winter seasons (Fig.6). In addition to, similar congeners (PCB 28, PCB 52, PCB 101) were found to be primary congeners for particle phase.

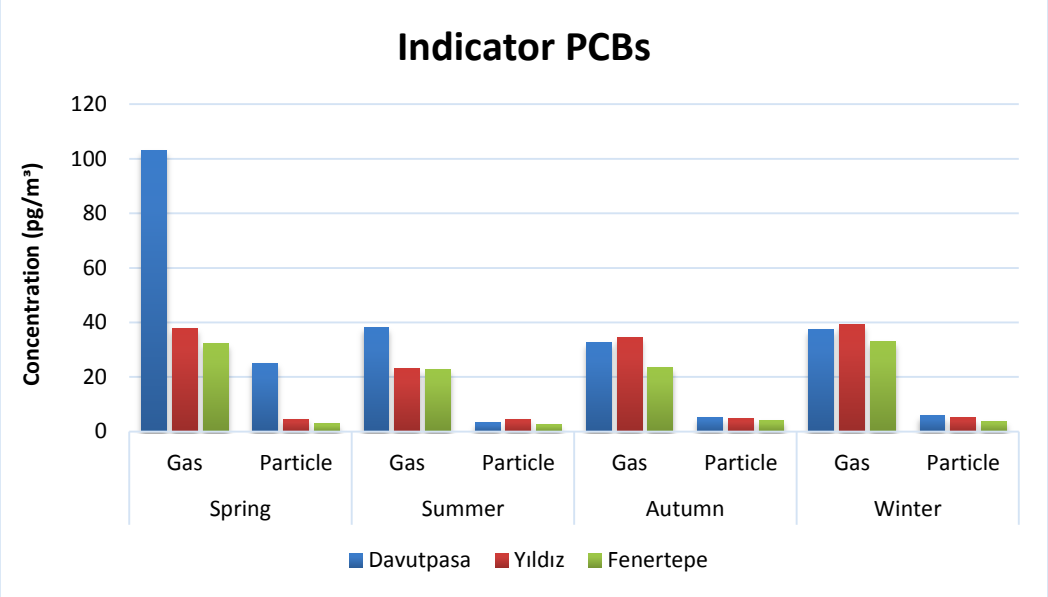

Fig.5 Seasonal variation of gas/particle partitioning of indicator PCB_s

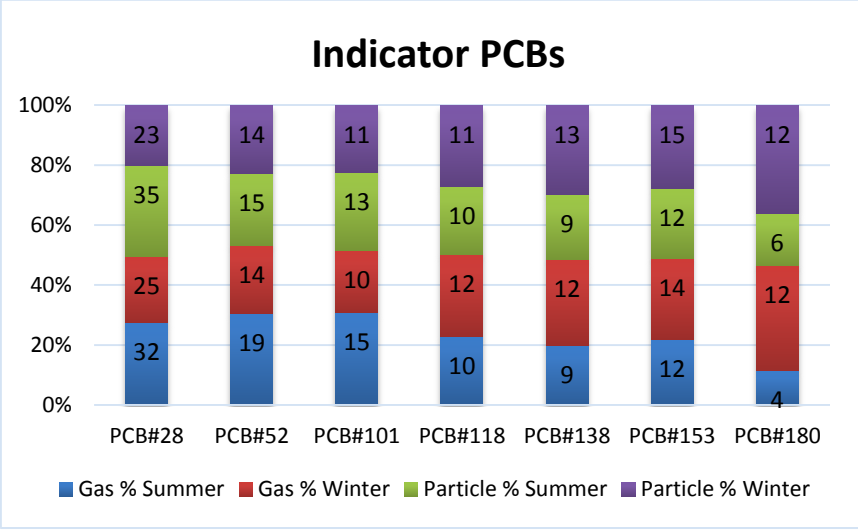

Fig.6 Gas/particle distribution of indicator PCB_s congeners

EFFECT OF TPM CONCENTRATION ON GAS/PARTICLE DISTRIBUTION

TPM concentration and physico-chemical properties of particles and congeners are important factors that are affecting the gas/particle distribution of semi volatile organic compounds. Average TPM concentrations for Davutpasa, Yıldız and Fenertepe sampling stations were detected to be 79, 78 and 44 μg m^{-3}, respectively. The values for Davutpasa and Yıldız are higher than the 24 h limit value (50 μg m^{-3}) reported by WHO (2005). The correlation coeffcient was detected to be R=0.46 between the total particle matter and particle phase dl-PCB, considering all sampling results. High correlation was found for winter (R=0.47, R^2=0.22) and summer seasons (R=0.46, R^2=0.21). This could be explained by the increased adsorption of the dl-PCBs on the particle matter due to the low ambient air temperature in winter season. TPM and dl-PCB_s may be emitted together and simultaneously from the source or sources during the summer season. For example, high correlation coefficient (R=0.8, R^2=0.65) was determined for Fenertepe sampling station during the summer season. As TPM concentration increases, the surface area for adsorption of the compounds are also increases. Thus, particle phase of the compounds can be rised. Therefore it has been estimated that TPM and dl-PCB_s were emitted together and simultaneously from the source or sources at Fenertepe sampling station. Medical waste incineration plant and gasification plant which are located at respective distances of 8 km NE and 12 km SE from Fenertepe sampling station. Low correlation (R=0.22) was found between the total particle matter (TPM) and non dioxin like PCB_s in the particle phase. Although high positive correlation was found (R=0.74, R^2=0.55) in summer season, there was no correlation in winter season. As a result different correlation coefficient were found between the total particle matter (TPM) and particle phase PCB_s conentrations. Different correlation coefficients can be explained by various factors including total particle matter concentration, properties of particle matter (carbon content and chlorination level) and vapor pressure of organic compounds. High positive correlations were found between UV (R=0.1), solar radiation (R=0.1), temperature (R=0.15), and dl-PCB concentrations while low negative correlations were found for precipitation (R=-0.1), wind rate (R=-0.13) and atmospheric pressure (R=-0.1). Similar to the dl-PCB_s, positive low correlations were found between the UV (R=0.1), SR (R=0.2) and indicator PCB concentrations while low negative correlations were found for wind rate (R=-0.24) and atmospheric pressure (R=-0.3). Determining low negative correlation for wind rate can be explained by the removal of particle phase PCB compounds via dry depositon mechanism. Bidleman (1988) reported that the highly chlorinated congeners can be deposited via dry and wet deposition mechanisms. Determination of low negative correlation for atmospheric pressure can be explained by desorption of PCBs from reservoir areas due to the high atmospheric temperature and low atmospheric pressure. However low regression coefficients (R^2<0.1) showed that the correlation between the meteorological parameters and PCB concentration are affected by several factors such as properties of the organic compounds and particle matter, seasonal variation etc.

CONCLUSION

In this study, gas/particle partitioning of the dl-PCB and indicator PCB compounds was investigated at three different areas in the metropolitan city of Istanbul. Dl-PCBs and non dl-PCB_s showed a tendency to be existed in gas phase at higher percentages (77% for dl-PCB and 90% for non dl-PCB). Average gas phase ratio of dl-PCB compounds increased significantly during summer season. Indicator PCB compounds did not show significant seasonal variation unlike dl-PCB compounds. The positive low correlations were found between the concentrations and temperature and SR, while negative low correlations were found for wind rate and atmospheric pressure.

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