# STATISTICAL ANALYSIS OF ATMOSPHERIC POLYCHLORINATED BIPHENYL CONCENTRATIONS AT TWO URBAN LOCATIONS

by

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#### **Abstract of the Thesis**

# STATISTICAL ANALYSIS OF ATMOSPHERIC POLYCHLORINATED BIPHENYL CONCENTRATIONS AT TWO URBAN LOCATIONS

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Atmospheric deposition of persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) is an important source of contamination to many U.S. water bodies, including the Delaware River and Lake Michigan. In this thesis, data on concentrations of PCBs in gas and aerosol phases in Chicago, IL and Camden, NJ were analyzed to identify factors that affect atmospheric PCB concentrations in urban areas.

In order to investigate factors driving atmospheric PCB concentrations in Camden, NJ, concentrations of gas and particle-phase PCBs, meteorological parameters (temperature, barometric pressure, wind speed, wind direction, relative humidity, and solar radiation) and three criteria pollutants (ozone, carbon monoxide, and nitrogen oxides) measured during 2000-2003 were analyzed by multi-parameter linear regression. The well-known temperature dependence of gas-phase PCBs was evident (average  $R^2 = 0.62$ ; p < 0.05). Gas phase PCB concentrations displayed weak but significant relationships with ozone, wind speed, relative humidity and solar radiation (p < 0.05).

When paired with temperature, only wind speed, relative humidity and/or solar radiation produced significant relationships (p < 0.05). Solar radiation and humidity are themselves negatively correlated (p < 0.05), with relative humidity seeming to be the stronger of the two predictor variables.

An advanced factor analysis method, Positive Matrix Factorization (PMF), was used to identify the dominant source types of polychlorinated biphenyls (PCBs) in the atmosphere of Chicago, IL. PMF identified 6 factors. When compared to Aroclor compositions, only factor 1 (which comprised 23% of the PCB mass in the data set) was clearly identifiable as Aroclor 1242. Although PCB levels in most environmental compartments are declining gradually, with half lives on the order of 5 to 20 years, atmospheric PCB concentrations in Chicago displayed a marked increase starting around 2005. Results demonstrate this increase was due to factor 4 (which appears to be a mixture of Aroclors 1248/1254,  $R^2 = 0.674$ ), and factor 5, which resembled Aroclor 1254 ( $R^2 = 0.731$ ). Factors 4 and 5 comprised 24% and 16% of the total PCB mass in the data set, respectively. Factors 3 and 6 displayed an exponential decay with half lives of 3.0 and 5.8 years, respectively.

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## **Chapter 1**

#### **General Introduction**

Polychlorinated biphenyls (PCBs) are well known for their persistence within various compartments of the environment. First introduced during the early part of the 1900s, PCBs were manufactured as mixtures under various trade names and are chemically and thermally stable. In the U.S., the vast majority of PCBs were manufactured by Monsanto and sold under the trade name "Aroclor" followed by a number that usually indicated their chlorine content. For example, Aroclor 1242 contained 42% chlorine by weight. Their stability made them ideal for use as dielectric fluids in capacitors and transformers, though they have also been used in items such as paints, dyes, carbonless copy paper, and plastics, as well as within liquids responsible for heat transfer and hydraulics applications [1].

An individual PCB molecule consists of a biphenyl skeleton with a maximum of ten sites at which chlorine substitution can occur, which allows for 209 possible chlorination patterns and 209 individual PCB congeners that may be present in the environment. Of these, only about 90 were prevalent in the Aroclors and are usually measurable in environmental samples. PCBs can be further categorized into "homologues" based upon the number of chlorine atoms present on the biphenyl structure [1].

Open use and manufacture of PCBs in the U.S. were banned in the 1970s. PCBs are still used in closed applications such as transformers, even today. Despite the ban,

present-day concentrations of these compounds in water and sediment are still high enough to exceed applicable standards, particularly in urban areas. PCBs are semi-volatile, allowing for the occurrence of atmospheric transport and subsequent deposition to locations other than the source region [1]. Atmospheric PCB concentrations are higher in industrialized, urban environments as opposed to rural areas [2-8]. PCBs are hydrophobic and tend to accumulate in soils, sediments and fatty tissue. Due to their lipophilic properties, biomagnification of PCBs can occur as contaminated organisms are consumed, leading to elevated PCB levels in aquatic organisms [9, 10]. In order to protect human health and prevent exposure or consumption of PCB contaminated biota, fish consumption advisories have been issued for areas where concentrations of PCBs in fish are exceptionally high [1, 10-13].

The Clean Water Act (CWA) requires the implementation of total maximum daily loads (TMDLs) in order to address the input of any contaminant that exceeds the applicable Water Quality Standards (WQSs) for any surface water body in the U.S. The purpose of a TMDL is to quantify the amount of a contaminant that can enter into a water body without surpassing the WQS. Water quality models that utilize estimated contaminant loadings from various sources are an important part of calculating TMDLs. Polychlorinated biphenyls are one class of contaminant that exceeds WQS in hundreds of water bodies throughout the U.S. PCB TMDLs have been promulgated for the Schuylkill, Susquehanna, Ohio, and tidal Potomac rivers, as well as the San Francisco Bay [10, 12-16]. In 2000, the TMDL process was undertaken in the Delaware River, where PCB concentrations exceed the 7.9 pg L<sup>-1</sup> WQS, put in place by the Delaware River Basin Commission (DRBC), by an order of magnitude [1]. The analysis by the

DRBC demonstrated that atmospheric deposition alone exceeds the total TMDL for Zones 2 through 5 of the Delaware River [4].

Research has shown that atmospheric deposition of PCBs is an important source of contamination, particularly in water bodies influenced by urban areas, such as the Delaware River and Lake Michigan. Long-term monitoring of PCBs has been ongoing in the areas adjacent to these water bodies, and high levels of PCBs have been reported [4, 6, 17]. Meteorological factors can affect the behavior of PCBs in the atmosphere. Relationships between PCBs and air temperature are well known [4, 18-27]. The effects of other variables such as wind speed, wind direction, and relative humidity on PCB concentrations and fluxes have also been examined in order to characterize the behavior of PCBs [17, 22, 28, 29].

Zhang et al. [17] detected a significant relationship (p < 0.05) between total gasphase PCB concentrations and wind direction in Chicago, IL. This relationship was strengthened by the inclusion of temperature. Similar relationships were seen at sites surrounding Lakes Erie, Michigan and Superior. Hillery et al. [22] reported a significant effect of temperature on total gas-phase PCB concentrations. The relationship improved slightly with the addition of wind speed and wind direction. In Birmingham, United Kingdom the effect of meteorological factors on PCB concentrations was investigated for ΣPCBs and also for individual congeners/homologue groups. Harrad and Mao [28] reported significant correlations between wind speed, wind direction and relative humidity on an individual congener basis, whereas Currado and Harrad [29] found that only wind direction had a significant impact on the lesser chlorinated congeners. In this thesis, a similar analysis of atmospheric PCB concentrations, wind direction, wind speed,

relative humidity, barometric pressure and temperature data for Camden, NJ, was performed. In addition, ozone, solar radiation, carbon monoxide and nitrogen oxide data were included in the data set to determine whether these variables, which are indicators of the photochemical reactivity of the atmosphere, had any effect on atmospheric PCB concentrations.

Another urban location where atmospheric PCB concentrations are elevated [8, 38, 39] and atmospheric deposition to the adjacent water body is an important source of PCBs is Chicago, Illinois. Dry particle fluxes of PCBs have been reported as being up to 3 orders of magnitude higher near Chicago than in remote regions [40]. Hafner and Hites [41] have used the Potential Source Contribution Function to identify the city of Chicago as a potential source of PCBs to Lake Michigan. Other studies have observed increases in PCB concentrations over the lake when winds were coming from the Chicago area [8, 17].

Long-term monitoring networks, such as the Integrated Atmospheric Deposition Network (IADN), allow for the characterization of temporal trends resulting from PCB contamination. IADN is a joint venture between the United States and Canada formed in November 1990 to address the loadings of persistent organic pollutants (POPs) to the Great Lakes. The U.S. sampling stations have been operated by Indiana University under the supervision of the United States Environmental Protection Agency's Great Lakes National Program Office. Monitoring occurs at five Master Stations, one for each of the Great Lakes, to characterize the regional background without the influence of local sources. In addition to the Master Stations, satellite sites are positioned in areas where urban influences can be examined [42].

Positive matrix factorization (PMF) has gained widespread application as a method of source apportionment in areas of air resource management [30-33] and has been used to identify types of PCB sources in the Philadelphia region in support of the Delaware River TMDL [34-36]. An advantage of PMF over other source apportionment tools is the ability to handle missing and below detection limit data [37].

This thesis is comprised of two parts, both of which aim to further the understanding of urban areas well known for their PCB contamination. Chapter 2 utilizes statistical analysis software to describe the relationship between numerous meteorological variables and atmospheric PCB concentrations in Camden, NJ. Chapter 3 uses a robust source apportionment tool, PMF, along with an extensive dataset in an attempt to identify the types of sources in Chicago, IL that are contributing to high levels of PCBs in Lake Michigan.

Gas and particle-phase PCB measurements, as well as earlier temperature, relative humidity, barometric pressure, wind speed and wind direction data for Camden, NJ were obtained through the New Jersey Atmospheric Deposition Network (NJADN). For about three years from 2005-2008, the candidate was partially responsible for sample collection and analysis for the NJADN, processing hundreds of samples for the network. Hourly ozone, carbon monoxide, nitrogen oxides, and solar radiation data were provided by the New Jersey Department of Environmental Protection. Chicago gas-phase PCB measurements were made available by the IADN.

The first section of this work will focus on using statistical analysis software (SAS) to perform simple linear and multiple linear regression analyses in order to determine possible interactions between PCB concentrations measured in Camden, NJ,

and meteorological variables that may be affecting fluxes of PCBs to the Delaware River.

The second part of this thesis will utilize PMF to identify the types of sources in the Chicago, IL atmosphere that may be influencing the gas-phase PCB concentrations reported for Lake Michigan.

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#### Chapter 2

The Influence of Meteorological Parameters on the Concentration of Polychlorinated Biphenyls in the Atmosphere of Camden, NJ

#### Abstract

Particle- and gas-phase concentrations of polychlorinated biphenyls (PCBs) were measured in Camden, NJ as part of the New Jersey Atmospheric Deposition Network (NJADN) in support of the effort to calculate PCB Total Maximum Daily Loads (TMDLs) for zones 2 through 5 of the Delaware River. These measurements show that PCB levels in Camden are about 20 times higher than in nearby suburban areas, such as Washington Crossing, NJ. In order to investigate the factors driving the atmospheric PCB concentrations in Camden, a data set containing concentrations of PCBs in the gas and particle phases, meteorological parameters, and concentrations of three criteria pollutants measured during 2000-2003 was analyzed by multi-parameter linear regression. Meteorological data (temperature, barometric pressure, wind speed, wind direction and relative humidity) were taken from the nearby National Weather Service site. Additional parameters (ozone, carbon monoxide, nitrogen oxides, and solar radiation) were obtained from measurements taken by the New Jersey Department of Environmental Protection (NJDEP) at a nearby site. Linear regression analysis was performed using statistical analysis software (SAS version 9.1). Regressions carried out using the particulate phase PCB concentrations were not significant for any of the atmospheric variables (at p < 0.05). It is widely recognized that gas-phase PCB concentrations are a function of temperature via the Clausius-Clapeyron equation, and

this relationship was evident in the data set (average  $R^2$  of 0.62; p < 0.05). Even without correcting for temperature, gas-phase PCB concentrations displayed weak but significant relationships with ozone, wind speed, relative humidity and solar radiation (average  $R^2$  values were 0.12, 0.16, 0.13 and 0.09, respectively and p < 0.05 for more than half of the total congener list). Multiple regressions that included the effect of temperature in addition to wind speed, relative humidity and/or solar radiation also produced significant relationships (p < 0.05); however, solar radiation and humidity are themselves negatively correlated (p < 0.05). Relative humidity is the stronger of these two variables due to the higher average  $R^2$  value and higher occurrence of significant slope coefficients.

#### **Introduction:**

In many urban systems, PCB concentrations exceeding the federal water quality standard have led to efforts under the Clean Water Act to establish Total Maximum Daily Loads (TMDLs) for these impaired water bodies. The purpose of the TMDL is to specify a maximum amount of pollutant that can be discharged into a body of water from all contributing sources while allowing said water body to attain water quality standards. The Delaware River Basin Commission (DRBC) developed a Total Maximum Daily Load (TMDL) of 380 mg/d for PCBs in zones 2 through 5 of the Delaware River. PCB TMDLs have also been developed for the Schuylkill River, San Francisco Bay, and the tidal waters of the Potomac River [1-7]. Table 2.1 provides a summary of implemented TMDL values for these water bodies.

The Delaware River PCB TMDL has been a phased effort to identify PCB sources along the river and to lower the ambient PCB concentrations, which currently

exceed the applicable water quality standards which are 44.4 pg/L for zones 2 and 3, 44.8 pg/L in zone 4 and 7.9 pg/L in zone 5 [5]. Recently, actions have been taken to amend the applicable water quality standard for PCBs in the entire length of the Delaware River to 16 pg/L. This effort would include Delaware Bay, which is currently designated as zone 6 and has a water quality standard of 64 pg/L (www.state.nj.us/drbc). Several monitoring networks have been established to characterize the effects that urban areas have on nonurban regions regarding contamination and atmospheric transport. The Integrated Atmospheric Deposition Network (IADN) was created to understand atmospheric deposition along the Great Lakes. Monitoring efforts performed through IADN have been successful in illustrating that PCB concentrations in urban Chicago are higher than those in rural areas and may contribute to higher concentrations measured in nearby coastal areas [8-11]. In addition, the Atmospheric Exchange Over Lakes and Oceans Study (AEOLOS) was a short term sampling endeavor conducted to address the issue of urban concentrations influencing coastal areas within Chicago-Southern Lake Michigan and the Baltimore Harbor-Northern Chesapeake Bay [9, 12-20].

The New Jersey Atmospheric Deposition Network (NJADN) focused on atmospheric concentrations of PCBs, PAHs, organochlorine pesticides, nutrients and trace metals at sites across New Jersey, Pennsylvania, and Delaware [21-25]. PCB concentrations measured at seven NJADN sites on the Delaware River were used to estimate atmospheric loads into the river as part of the Delaware River PCB TMDL process. Concentrations of gas-phase PCBs at the Swarthmore, PA, and Camden, NJ, monitoring sites averaged 3300 pg/m³, and were much higher than concentrations found at the remaining five sites [22]. The Camden sampling site is located within Zone 3 of

the Delaware River and is considered urban (Figure 2.1). Based on the elevated gasphase PCB concentrations at the site and evidence that contamination in urban areas can affect coastal regions, trends were examined between PCB concentrations in Camden and meteorological parameters so that the cycling of PCBs in the Delaware River Bay can be better understood.

Due to the recalcitrance of PCBs, the compounds are not likely to break down easily within the environment. Aerobic and anaerobic biodegradation does occur, but these processes are very slow [26]. Recently, research has verified that gaseous PCBs react with the hydroxyl radical (OH) at significant rates [27-33]. Approximately 90% of PCBs present in the atmosphere are found in the gas phase, with the remaining 10% (mainly the heavier PCB congeners) being sorbed to particles [22, 24]. This distribution between the gas and particle phase means that PCBs fall into the class of semi-volatile organic compounds (SVOCs). Anderson and Hites [27] measured rate constants for the reactions of 14 PCB molecules with the OH radical. Second-order rate constants ranged from  $5.0 \times 10^{-12} \text{ cm}^3 \text{s}^{-1}$  to  $0.4 \times 10^{-12} \text{ cm}^3 \text{s}^{-1}$  for 3-chlorobiphenyl and  $2.2^{\circ}, 3.5^{\circ}, 6$ pentachlorobiphenyl, respectively. They estimated the atmospheric lifetimes of PCBs due to reactions with OH ranges from days for biphenyl to 34 days for pentachlorobiphenyl. This and other research demonstrated that the rate of reaction between PCBs and the OH radical decreases as the number of chlorine substituents increases [27, 29, 30]. Assuming a global average OH concentration of 9.7 x 10<sup>5</sup> molecules cm<sup>-3</sup>, Anderson and Hites calculated a flux equal to 16 μg·m<sup>-2</sup> yr<sup>-1</sup> of PCBs being removed from the atmosphere through the reaction with the OH radical. Based on

this, they argue that reactions with the OH radical are the largest sink for PCBs worldwide.

Since these laboratory studies, a few field studies have presented evidence for destruction of PCBs by the OH radical during atmospheric transport. Totten et al. [30] observed a decrease in PCB concentrations at three urban sites during daytime and proposed that it was caused by reactions with OH radical during tropospheric transport. Second-order rate constants calculated from this decrease were consistent with laboratory measurements [27]. Mandalakis et al. [29] computed the lifetimes of PCBs 8 and 110 utilizing atmospheric measurements in Finokalia, Greece. They calculated that in tropical/subtropical regions the lifetimes were 10 days for PCB 8 and 20 days for PCB 110.

Attack by OH radical is the primary pathway of removal for many volatile or semi-volatile organic compounds present in the atmosphere. The OH radical is formed mainly through the photolysis of ozone (O<sub>3</sub>), via the following reaction:

$$O_3 + hv (\lambda \le 336 \text{ nm}) \rightarrow O(^1D) + O_2$$
  
 $O(^1D) + H_2O \rightarrow 2OH$ 

The lifetime of the OH radical within the atmosphere is very short (on the order of seconds) and its formation is dependent on the presence of sunlight. As a result, reactions of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) with the OH radical occur primarily during the day [34, 35]. Thus it expected that gas-phase PCB concentrations will be measurably lower during periods of high photochemical activity. In addition, the amount of tropospheric O<sub>3</sub> present has a direct effect on OH concentrations. Increased levels of O<sub>3</sub> will result in greater OH production

within the atmosphere, which will lead to larger decreases of OH reactive species [36]. In urban areas, higher levels of  $O_3$  in the atmosphere are associated with  $NO_x$  and VOC emissions originating from industrial sources and automobiles [37, 38].

Measurement of OH radical concentrations in the troposphere is difficult [35, 39]. Determination of OH radical concentrations primarily involves estimation utilizing tracer species, OH radical scavengers, or complicated predictive models [35, 39-43]. Such models use concentrations of other photochemically reactive species, such as  $O_3$  and  $NO_x$ . These species are easily measured and routinely monitored at various locations, including areas in the vicinity of NJADN samples sites. Therefore in the present work, the relationships between atmospheric PCB concentrations and concentrations of  $O_3$  and  $NO_x$  were examined to look for evidence of reactions of PCBs with OH radical.

The main purpose of this study was to investigate the relationship between gasphase PCB concentrations and atmospheric conditions previously recorded in Camden, NJ. Correlations between gas-phase PCB concentrations and nine variables were investigated: ozone (O<sub>3</sub>), carbon monoxide (CO), nitrogen oxides (NOx), wind direction (WD), wind speed (WS), temperature (T), relative humidity (RH), barometric pressure (BP), and solar radiation (SR). Linear regressions were performed using SAS version 9.1 in an attempt to better understand how atmospheric conditions might predict the behavior of PCBs found in the atmosphere of an urban location.

#### **Experimental Section:**

#### Data collection:

Data from the Camden site was used in this investigation because it is the most urban of the NJADN sites and has one of the longest data histories. The site is located on the roof of the Paul Robeson Library on the Rutgers University Campus in Camden, NJ. Full data can be found in the Final Report for the NJADN project [44] and is summarized in Totten et al. [24]. Meteorological data was obtained from the NOAA National Weather Service meteorological station located at the Philadelphia International Airport. Ozone, carbon monoxide, solar radiation, and NO<sub>x</sub> measurements were taken 3 miles from the Paul Robeson Library at the Camden Lab, located on Davis Street in Camden, NJ, and was provided by the New Jersey Department of Environmental Protection (NJDEP). Hourly averages of ozone  $(O_3)$ , carbon monoxide (CO), temperature (T), wind speed (WS), wind direction (WD), barometric pressure (BP), solar radiation (SR) and relative humidity (RH) were obtained for July 1999 through August 2002. NO<sub>x</sub> hourly averages were also obtained, however, the data set did not begin until June of 2000. As a result, data for July 1999 through May 2000 for all other parameters was not included in any of the statistical analysis. The final data set included the June 2000 through August 2002 time period. PCB samples were collected from 9 am to 9 am. Therefore, for comparability, the hourly averages reported for ozone, CO, solar radiation and NOx were used to calculate each variable's daily averages over the same 24-hour period.

#### Sample Collection and Analysis:

Full details of sampling and analysis are available in the Final Report for the NJADN project [44] and are summarized in Totten et al. [24]. The PCB data were collected as part of the NJADN study during 1999- 2002 at Camden, NJ. Air samples were collected every 12 days for 24 hours using a high volume air sampler (Tisch Environmental, Village of Cleves, OH), calibrated with a flow rate of ~0.5 m³min⁻¹. The particulate and gas phases were collected using quartz fiber filters (QFF; Whatman) and polyurethane foam plugs (PUF), respectively. In preparation for sample collection, the QFFs were baked at 450°C for 24-hours in aluminum foil envelopes followed by a 24 hour equilibration period in a temperature and humidity controlled chamber. The weights of the QFFs were recorded. The filters were stored in the aluminum foil envelopes at 4°C until sample collection. PUFs were cleaned via soxhlet extraction; 24 hours in acetone followed by 24 hours in petroleum ether. The PUFs were then dried in a dessicator for 48 hours then stored, individually, in precombusted glass jars at room temperature until sampling. Upon collection, samples were taken back to the lab and stored at 4°C.

Prior to soxhlet extraction, the QFFs were left in the temperature and humidity controlled chamber for 24-hours to re-equilibrate. The post-weight of each QFF was obtained and, along with the pre-weights, the total suspended solids (TSS) associated with each sample was calculated. The QFFs were then returned to the freezer until extraction. Prior to use, all glassware was rinsed with hexane in triplicate. Glassware was rinsed with Milli-Q water and baked at 450°C over night after use.

The QFFs and PUFs were extracted in dichloromethane (DCM) and petroleum ether, respectively, for 24-hours using a soxhlet/condenser/round bottom flask apparatus. Prepared PCB and PAH surrogate standards (PCBs 14, 22, 65, 166, and PAHs d<sub>10</sub>-

anthracene, d10-fluoranthene, d10-benzo[e]pyrene) were injected onto the samples before addition of the solvent. Upon completion of the extraction, the round bottom flasks containing the solution of sample and solvent underwent rotary evaporation (Büchi Model RotoEvaporator111). Samples were concentrated to about 2 ml, the solvent was switched to hexane, and extracts were transferred to 12 mL amber vials. Next the samples were concentrated to about 0.5 mL under a gentle stream of purified N<sub>2</sub> gas using a nitrogen-evaporator (Organomation Associates 111) in preparation for PCB and PAH fractionation by deactivated aluminum oxide (Al<sub>2</sub>O<sub>3</sub>).

Aluminum oxide (Neutral Alumina, Brockman Activity I, A950-500, 60-325 mesh: Fischer Scientific) was baked at 550°C for 24 hours and deactivated with 3% deionized water by weight. The samples were injected onto precombusted columns containing the deactivated aluminum oxide and eluted with 13 mL of hexane to separate out the PCB fraction. The PAHs were eluted with 15 mL of a 2:1 DCM/hexane mixture. Both fractions were concentrated under a gentle stream of nitrogen gas to about 1 mL, transferred to 2 mL amber vials with hexane and stored at 4°C until analysis. The PCB fraction was injected with internal standard (PCBs 30 and 204) and analyzed.

PCBs were analyzed using an HP 6890 gas chromatograph equipped with 63Ni electron capture detector using a 60-m 0.25 mm i.d. DB-5 (5% diphenyl-dimethyl polysiloxane) capillary column with a film thickness of 0.25 µm. The temperature program was as follows: 70 °C to 180 °C at 7 °C/min; 180 °C to 225 °C at 1.05 °C/min; 225 °C to 285 °C at 5.75 °C/min; 285 to 300 at 11.50 °C/min; hold 300 °C for 8 minutes. Sixty peaks representing 93 congeners were quantified. Calibration standards were analyzed with field samples and contained all congeners of interest. In instances where

coelution was observed, the most abundant congener in the Aroclors was used to assign the peak to a homolog group. For quality control purposes, field blanks, laboratory blanks and matrix spikes were included at a rate of 15%, 10% and 5% of samples, respectively.

#### Statistical Analysis:

PCB congener concentrations were transformed using the natural logarithm for both particle and gas phases (ln C<sub>particle</sub> and ln C<sub>gas</sub>, respectively) for use in linear regression analysis. Temperature measurements were converted into units of Kelvin and the inverse was taken. Wind direction (WD) data was originally reported in units of circular degrees. As in other studies [14, 45-49] WD was represented in the regression equations by the sine and cosine of the daily averages in order to account for the vector component of the data, with the sine and cosine representing the east-west and the north-south components, respectively. The WD values reported in circular degrees were averaged over the corresponding 24 hour period. The sine and cosine of the resulting averages were taken and used in the regression analysis. Data points for the remaining variables (ozone, barometric pressure, solar radiation, relative humidity and wind speed) did not need to be transformed and were, therefore, incorporated into the regressions in the format in which each was reported. The following relationship was examined:

$$ln C = a_0 + a_1 X$$
(2.1)

where C is the concentration of PCBs in either the gas or particle phase, and X represents  $O_3$ , CO,  $NO_x$ , WS,  $cos(WD_{ave})$ ,  $sin(WD_{ave})$ , 1/T, BP, RH or SR.  $O_3$ , CO,  $NO_x$ , and SR are each involved in photochemical reactions. It was expected that elevated levels of

each would result in decreased concentrations of PCBs in an urban atmosphere such as Camden.

#### **Results and Discussion**

Totten et al. [22] have previously reported that average gas- and particle-phase concentrations of PCBs for the Camden area are 3300 and 180 pg/m³, respectively. Regression coefficients were computed on a sum and individual congener basis. Complete tables containing regression parameters can be found in Appendix I. Results from the simple linear regressions for both ln C<sub>gas</sub> and ln C<sub>particle</sub> can be found in Tables I- 1 and I-2, respectively.

### Particle phase PCB concentrations

Tables I-1 and I-2 summarize the linear regression results obtained when the  $\ln C_{gas}$  and  $\ln C_{particle}$  was regressed against  $O_3$ , CO, NOx, WS, WD, 1/T, BP, RH and SR. The  $\ln C_{particle}$  produced weak relationships with all nine variables  $(O_3, CO, NOx, WS, WD, 1/T, BP, RH and SR)$ , none of which were significant (at p < 0.05, Table I-2). The poor correlations resulting from analysis of the  $\ln C_{particle}$  could be explained through incomplete equilibrium of gas/particle partitioning. In an urban site such as Camden, PCB emissions are fresh and do not have time to equilibrate. Furthermore, particles have much shorter lifetimes in the atmosphere than gases, and are more likely to be generated locally; whereas the gas phase PCBs could have been generated within a ~10 km radius and transported to the site by wind.

#### **Temperature**

The relationship between  $\ln C_{gas}$  and the inverse of temperature was strongest and most significant, with  $R^2$  values ranging from 0.34 to 0.89 for IUPACs 18 and 203+196, respectively. All regressions with 1/T resulted in p-values < 0.05. Table I-1 shows that the relationship between the sum of gas phase PCBs and inverse temperature (1/T) resulted in a stronger correlation with an  $R^2$  of 0.601 (p < 0.001), than the sum of particle phase PCBs ( $R^2 = 0.033$ , at p < 0.05, Table I-2).

The temperature dependence of PCBs can be explained through the Clausius-Clapeyron equation:

$$\ln P = -\frac{\Delta H_{SA}}{RT} + constant \tag{2.2}$$

where P is the partial pressure in atm, T is the temperature in K,  $\Delta H_{SA}$  represents the surface-air enthalpy of vaporization in kJ/mol and R is the universal gas constant (8.315 ×  $10^{-3}$  kJ/mol·K) [50]. In situations where the compound is expressed as a concentration, the following relationship applies:

$$\ln C_{gas} = -\frac{\Delta H_{SA} + RT_{av}}{R} \times \frac{1}{T} + constant$$
 (2.3)

where the term RT<sub>av</sub> is added to the  $\Delta$ H<sub>SA</sub> term, and T<sub>av</sub>, expressed in K, refers to the average temperature [50]. The RT<sub>av</sub> term was subtracted from the slopes calculated by SAS to obtain  $\Delta$ H<sub>SA</sub> in kJ/mol. For this study, the computational values of  $\Delta$ H<sub>SA</sub> symbolize the enthalpy of PCB transfer between surface and air.  $\Delta$ H<sub>SA</sub> values calculated using the slopes obtained through regression analyses were comparable with previously reported enthalpies of air-surface exchange, enthalpies of vaporization and enthalpies of plant-air exchange,  $\Delta$ H<sub>SA</sub>,  $\Delta$ H<sub>VAP</sub>, and  $\Delta$ H<sub>PA</sub>, respectively (Table I-5).  $\Delta$ H<sub>PA</sub> was derived

using the air/plant partition coefficient ( $K_{PA}$ ).  $K_{PA}$  was used to describe the dry gaseous deposition of PCBs and other SOCs to a plant's surface, in this case, ryegrass [56].

In general, the calculated  $\Delta H_{SA}$  values exhibited an upward trend with increasing molecular weight (Figure 2.2). This trend becomes more apparent when  $\Delta H_{SA}$  values are broken down into homolog groups (Figures 2.3 and 2.4). Figure 2.5 illustrates that the congener specific  $\Delta H_{SA}$  values computed for this study were generally lower than  $\Delta H_{VAP}$  measurements obtained in the laboratory [51-54]], as well as  $\Delta H_{SA}$  and  $\Delta H_{PA}$  values derived from atmospheric PCB concentrations measured elsewhere [48, 49, 55-58].

 $\Delta H_{SA}$  values computed here were plotted against  $\Delta H$  measurements previously reported for PCBs [48, 49, 51-58]. Figure 2.6 illustrates the relationship between  $\Delta H_{SA}$ calculated for this study and  $\Delta H_{VAP}$ ,  $\Delta H_{SA}$ , and  $\Delta H_{PA}$  values obtained from the literature. With the exception of  $\Delta H_{SA}$  values reported by Hoff et al. [59] for a rural site in Southern Ontario, all of the remaining data sets are significantly correlated (p<0.05) with those from Camden. The association between our enthalpies and those reported by Panshin and Hites [58], which were derived from concentrations measured at a contaminated site in Bloomington, Indiana, is weak (R<sup>2</sup>=0.26). Panshin and Hites found their enthalpies of vaporization to be in good agreement with those of Hoff et al. [59]. This is not surprising since the sites investigated by Hoff et al. and Panshin and Hites are characterized as rural. Panshin and Hites also reported significantly lower  $\Delta H_{SA}$  values, particularly for the higher molecular weight PCBs, when comparing their measurements to  $\Delta H_{VAP}$  values measured under controlled laboratory conditions by Falconer and Bidleman [58]. In general,  $\Delta H_{SA}$  values reported for Camden are more correlated with values of  $\Delta H_{VAP}(\mathbb{R}^2 \geq$ 0.70) obtained either through direct measurement of vapor pressure or quantitative

structure-property relationships (QSPR) [51-54]. When  $\Delta H_{SA}$  and  $\Delta H_{PA}$  values were derived using data obtained in the field, the measurements were limited to the congeners quantified at each sample site, decreasing the number of congeners available for comparison.

### Correlations with individual parameters (O<sub>3</sub>, CO, NOx, BP, WS, WD, RH, SR)

Regressions were also carried out between  $\ln C_{gas}$  and the nine variables (O<sub>3</sub>, CO, NOx, WS, WD, 1/T, BP, RH and SR). Only CO, NOx, WD and BP yielded correlations that were not significant (at p < 0.05) for more than half of the congeners. NO<sub>x</sub> is a major component in the formation of O<sub>3</sub>, which is in turn a major component of OH formation, suggesting that elevated levels of NO<sub>x</sub> and O<sub>3</sub> could be related to increased OH concentrations, and therefore larger decreases of  $\ln C_{gas}$ . The poor relationships between  $\ln C_{gas}$ , O<sub>3</sub> and NO<sub>x</sub> do not necessarily suggest that gas-phase PCB are not reacting with OH. Rather, the lack of correlation could merely suggest that O<sub>3</sub> and NO<sub>x</sub> are not good surrogates for OH radical concentrations. The lack of a significant relationship between  $\ln C_{gas}$  and WD could indicate that the sampling site in Camden, NJ, is situated in an area with sources coming from all directions.

## Temporal trends

Temporal trends were examined through expansion of the Clausius-Clapeyron equation yielding the following relationship:

$$\ln C_{gas} = a_0 + \left(\frac{1}{T}\right) + a_2 t \tag{2.4}$$

where t is the day relative to January 1, 1990. The  $\Sigma PCBs$  ( $R^2 = 0.61$ ) and individual congeners/coeluting groups ( $R^2$  ranged from 0.36 to 0.90) produced significant relationships (p < 0.05). The temperature coefficient for the  $\Sigma PCBs$  was -6896  $K^{-1}$  and ranged from -5399 to -14618  $K^{-1}$  (p < 0.05) for individual congeners/coelutors; while the time coefficient for  $\Sigma PCBs$  was -3.8 x  $10^{-4}$  days and ranged from -3.24 x  $10^{-3}$  to +1.17 x  $10^{-3}$  days, but was significant for only 5 of the 59 congener/coeluting groups (Table I-3). When compiling the complete dataset for analysis, data could only be included for those instances (i.e. sample dates) where values were available for all variables being considered in the regressions. The absence of data for several variables during specific time periods resulted in the exclusion of available data reported for those time periods in question. In the end, the dataset used for all analyses spanned from June 2000 through August 2002. A time interval of that length is not long enough to see temporal trends.

Correlations with individual variables  $(O_3, WS, WD, BP, RH, or SR)$  combined with temperature

The Clausius-Clapeyron was expanded again for 6 of the original variables:

$$\ln C_{gas} = a_0 + a_1(\frac{1}{T}) + a_2 X \tag{2.5}$$

where X is  $O_3$ , WS, WD, BP, RH, or SR. The inclusion of 1/T both increased the  $R^2$  values and lowered the p-values to a level of statistical significance for most of the parameters (Table I-5, a - f). For  $O_3$ , inclusion of 1/T in the regression raised the p-values (lowered the level of significance) of the  $O_3$  parameter so that it was significant for only a few congeners. This is due to the fact that  $O_3$  and 1/T are correlated (correlation coefficient = -0.51), with high  $O_3$  levels generally experienced in the hot summer months.

WS, RH, and SR remained statistically significant parameters (p<0.05) for the majority of congeners examined (Tables I-5, a - f).

The significance of wind direction on the atmospheric concentrations of PCBs has been observed in several studies [9, 14, 18, 60], but was not seen in this study. Hillery et al. [47] and Hoff et al. [59] also found that WD did not appear to have a significant correlation with PCB concentrations near Lake Superior or Egbert, Ontario, respectively. Currado and Harrad [49], however, did find a positive correlation between the cosine of WD and ln P for the sum of PCBs, and tri- and tetrachlorinated PCBs in Birmingham, UK. Brunciak at al. [46] observed that atmospheric concentrations of PCBs in New Brunswick, NJ increase when the winds were coming from the east.

The results of this study indicate that, in addition to the correlation between gasphase PCB concentrations and temperature, there is a significant relationship between  $\ln$  C<sub>gas</sub> and SR, WS and RH. This association was examined by, once again, expanding the Clausius-Clapeyron equation:

$$\ln C_{gas} = a_0 + a_1(\frac{1}{T}) + a_2WS + a_3RH + a_4SR$$
 (2.6)

where WS is the wind speed, RH is the percent relative humidity and SR is the solar irradiance (Table I-6). The average  $R^2$  of all 58 multiple regressions was 0.754 and all were significant (p < 0.05). In examining the regressions more closely, the coefficients for WS and RH were found to be significant for at least half of all congeners tested, while only two of the coefficients for SR exhibited significance (p < 0.05). The SR term was therefore removed and the following relationship was repeated for each of the congeners:

$$\ln C_{gas} = a_0 + a_1(\frac{1}{T}) + a_2WS + a_3RH \tag{2.7}$$

While the exclusion of SR did not drastically change the  $R^2$  (average  $R^2$  was equal to 0.750), congeners for which the WS and RH coefficients were not significant (at p < 0.05) when SR was a part of the model became significant (p < 0.05) when SR was removed (Table I-7). This is presumably due to the correlation between SR and RH, with high SR leading to low RH (see below for further discussion).

The coefficients for WS ( $a_2$ ) were negative, ranging from -0.034 to -0.228 and from -0.041 to -0.248 for multiple regressions run with and without the SR term, respectively. The negative sign indicates that PCB concentrations decreased with increasing WS, indicating a dilution effect. Average WS coefficients were -0.131  $\pm$  0.042 when SR was included in the relationship and -0.140  $\pm$  0.044 when SR was omitted. Twenty-eight of the 58 PCB congeners/coeluting groups resulted in WS coefficients that were statistically significant (p <0.05), and this number increased to 36 when the analyses were repeated without the SR variable. In both cases, the majority of the congeners/coeluting groups that exhibited significance towards the WS term were the lesser chlorinated congeners (Figure 2.7). When the model incorporated SR, the WS coefficients were slightly higher (less negative) than those calculated in the absence of SR.

The RH coefficients computed in this study were smaller in magnitude but positive in sign compared to those determined for WS. The positive sign indicates that PCB concentrations were higher on days with higher RH. Values of the RH coefficient ranged from 0.011 to 0.036 and averaged 0.018 in the presence of the SR term and from 0.004 to 0.019 with an average of 0.015 when SR was removed. The number of PCB congeners/coeluting groups that produced a statistically significant RH coefficient was 39

and the number increased to 44 in the absence of the SR variable. As was seen with WS, RH coefficients were generally higher when SR was excluded from the analysis. Unlike the WS term, in which most of the significant coefficients were associated with the lighter congeners, RH coefficients did not display any pattern or trend when plotted by IUPAC number (Figure 2.8).

The inclusion of the SR term resulted in slightly higher coefficients for WS and RH, but it also lowered the occurrence of statistically significant coefficients (p < 0.05). The 95% confidence intervals corresponding to the WS variable overlapped for the regression conducted with and without SR, while the confidence intervals related to RH did not. Thus, at the 95% confidence level, the RH coefficients obtained in the regression without SR were statistically different than those obtained when SR was included in the regression. The correlation seen between SR and RH coefficients is expected (correlation coefficient = -0.55). Increased levels of SR are associated with lower RH levels.

Previous studies have shown that wind speed can influence contaminant concentrations at some locations [12, 21, 38, 46, 61, 62]. The negative slope coefficients associated with WS in this study indicate a decrease in ln C<sub>gas</sub> as wind speed increases, suggesting a diluting effect. Totten et al. [21] found that a decrease in Jersey City, NJ PCB concentrations was correlated with an increase in wind speed. In addition, Bamford et al. [12] attributed lower gas phase concentrations of PCBs measured around Baltimore in February of 1997 to an occurrence of higher, more variable wind speeds. Brunciak et al. [46] reported that wind speed and gas phase PCB concentrations in a suburban area of NJ were significantly correlated, but when examining data collected at a coastal site in NJ during the same time period wind speed was not a significant variable.

The statistical relationship between RH and the gas phase concentrations of various persistent organic pollutants (POPs) has been investigated previously. In Birmingham, United Kingdom, a similar study used multiple regression analysis to identify relationships between atmospheric PCB concentrations, temperature and several other variables, including RH. In contrast to the results of this study, measurements taken in the UK show gas-phase PCB concentrations were not significantly correlated with RH [49]. A second set of atmospheric PCB concentrations was collected at the same site in Birmingham, UK and evaluated using the same method of multiple regression analysis. A positive relationship between RH and PCB concentrations was found to be significant, but only for data points corresponding to a wind speed of less than 4 m/s [48]. Raun et al. [63] found a negative correlation between RH and the concentrations of dioxins and furans in Houston, Texas. In addition, Hippelein and McLachlan [64] reported a strong correlation between RH and calculated soil/air equilibrium partitioning coefficients (K<sub>SA</sub>) for several PCB congeners. As RH decreased, K<sub>SA</sub> values increased indicating that as the soil dried, more PCBs were released back into the atmosphere.

The correlation between SR and RH seen in this study could be related to the drying of soil. Incoming solar radiation is important to the temperature of the soil surface. As solar radiation and soil temperature increase, evaporation of soil water is enhanced allowing for the transport of contaminants to the soil surface from below where they can volatilize into the atmosphere [65, 66]. Kurt-Karakus et al. [67] reported greater soil-to-air fluxes of DDT during spring months when observed solar radiation was higher. In another study, PCB volatilization from wet sediments was monitored over time as the sediments were allowed to dry under ambient conditions. PCB volatilization and water

loss were positively correlated. The rate of PCB loss from the sediment slowed significantly when water evaporation was complete suggesting that volatilization of PCBs is enhanced when evaporation is occurring from wet soils and sediments [68, 69]. Miskewitz et al. [70] similarly observed increased volatilization of PCBs from stabilized dredged material associated with the presence of solar radiation.

Prueger et al. [71] observed that volatilization fluxes of metolachlor followed the same diurnal pattern as solar radiation and that volatilization and solar radiation peak measurements generally coincided with one another on a daily basis. Correlations between metolachlor volatilization and relative humidity were also observed. It was speculated that the combination of dry soil surface, increasing atmospheric relative humidity and transport of soil water vapor from below the surface contributed to the correlations seen between volatilization and relative humidity. In general, metolachlor volatilization was at its highest during periods of warm, wet soil conditions.

In addition to affecting volatilization of chemicals from the soil, SR can influence reactions occurring in the atmosphere. Solar UV radiation is necessary for the formation of the OH radical, which reacts with PCBs [27-30, 39]. The photodegradation of PCBs has been effectively examined under simulated solar conditions, and usually in the presence of solvents or surfactants, which enhance degradation. Direct photolysis of PCBs occurs mainly via photolytic dechlorination [72-86]. Recently, Lores et al. [87, 88] used solid-phase microextraction (SPME) fibers as a means to monitor the photolysis of PCBs and identify photoproducts in a solvent-free environment. SPME fibers were also utilized to obtain photoproducts resulting from direct photolysis of aqueous solutions containing PCBs. Lesser chlorinated biphenyls were obtained as photoproducts in both

cases, confirming that SPME extraction is an effective method for examining PCB photolysis.

To better understand the results of the multiple regression analysis, the computed coefficients for each variable (WS, RH and SR) were regressed against log vapor pressure values obtained from Falconer and Bidleman, [54]. Of the three variables, WS is the only variable that exhibited a significant relationship with vapor pressure (R<sup>2</sup> = 0.336, p < 0.05). The data illustrate an inverse relationship between WS and log VP, where larger vapor pressures were correlated with lower (more negative) WS coefficients (Figure 2.9). If the effect of increased WS is to dilute the atmospheric concentrations of PCBs, then this physical process is expected to affect all congeners equally. The fact that WS affects different congeners differently suggests that dilution may not be the only or even the primary effect that WS has on the atmospheric concentrations of contaminants.

High molecular weight PCBs possessing lower vapor pressures are primarily associated with the particle phase and exit the atmosphere via wet or dry deposition. Dry deposition of particles occurs through gravitational settling, where as dry gaseous deposition takes place due to diffusion. Some studies have suggested that the dry deposition velocities of PCBs increase with chlorination [89, 90] and with wind speed [96, 97]. Therefore, the lighter congeners with a lower degree of chlorination will have lower dry deposition velocities than the heavier congeners at higher wind speeds [89, 90]. This would cause the concentrations of the lighter congeners to be less affected by wind speed than the heavy congeners, contrary to what is observed. Faster wind speeds also drive faster air/water exchange of pollutants such as PCBs [12, 14, 92-94]. Lower MW congeners have faster air/water exchange velocities, so in areas where the net direction of

air/water exchange results in gas absorption, faster wind speeds could result in more uptake of lighter congeners into the water phase, removing them from the atmosphere. However, in the Delaware River near Camden, the net direction of air/water exchange results in volatilization of PCBs from the water column [95].

Uptake rates of PCBs into passive air sampling media such as polyurethane foam is faster at higher winds speed and for lower MW congeners [96-101]. If this is true for other environmental compartments such as soil and vegetation, and if these other compartments are net sinks for gas-phase PCBs, then faster WS would result in faster uptake of low MW PCBs, resulting in lowered gas-phase concentrations [101-103].

## Conclusions:

The purpose of this study was to look for correlations between PCB concentrations and various atmospheric parameters to 1) characterize the behavior of PCBs in urban Camden, NJ and 2) detect reactions of PCBs with the OH radical through correlations of PCB concentrations with nitrogen oxide (NO<sub>x</sub>) compounds and ozone (O<sub>3</sub>). The lack of a significant negative correlation between O<sub>3</sub> levels and gas-phase PCB concentrations does not necessarily imply that OH radical reactions are unimportant for PCBs. Instead, NO<sub>x</sub> and O<sub>3</sub> may be poor surrogates for OH radical concentrations despite the fact that they are used as inputs for models developed to estimate OH concentrations [43]. Such models are complex and involve multiple feedback loops, which can lead to conditions in which elevated O<sub>3</sub> concentrations do not produce high OH radical concentrations. Other studies have suggested that reactions with OH radical may be unimportant in determining gas-phase PCB concentrations. Macleod et al. [104]

used a simple atmospheric box model to examine diel variations in gas-phase PCB concentrations and concluded that reactions involving OH radical are relatively unimportant when compared to other atmospheric variables such as mixing height or wind speed.

The number of data points used in the analysis was limited by the availability of NOx data for the Camden, NJ, area. Collection of this data did not begin until June of 2000. This resulted in a final range of only 2 years (2000-2002). The next step would be to perform the analysis using a much larger data set, either by rerunning the regressions with an updated Camden, NJ, PCB database or compiling NO<sub>x</sub> and O<sub>3</sub> in an area where PCBs have been measured for a longer time period (e.g. Chicago) and modifying the regression equations to include variables thought to have an impact on OH radical concentration.

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Table 2.1. Summary of previously implemented TMDLs.

Body of Water		TMDL (mg/day)	References
Delaware River	Zone 2	380 257	Fikslin and Suk, 2003
	Zone 3	18	
	Zone 4	57	
	Zone 5	48	
Delaware Bay	Zone 6	1,876	Fikslin and Suk, 2006
San Francisco Bay		27,397	CRWQCB, 2007
Total Tidal Potomac/Anacostia River		4,137	Haywood and Buchanan, 2007
Ohio River	River mile 40.0 to 161.7	15,222	ORSANCO, 2002
Schuylkill River	Mainstem	7,450	Tetra Tech, Inc., 2007
Susquehanna River		647	PADEP, 1999

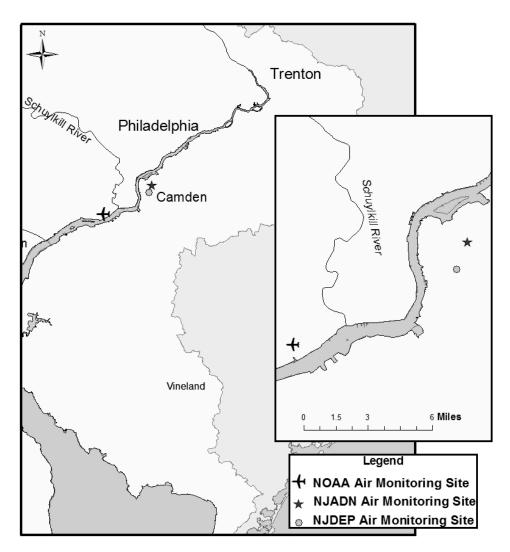


Figure 2.1. Location of sampling sites in the Philadelphia/Camden area.

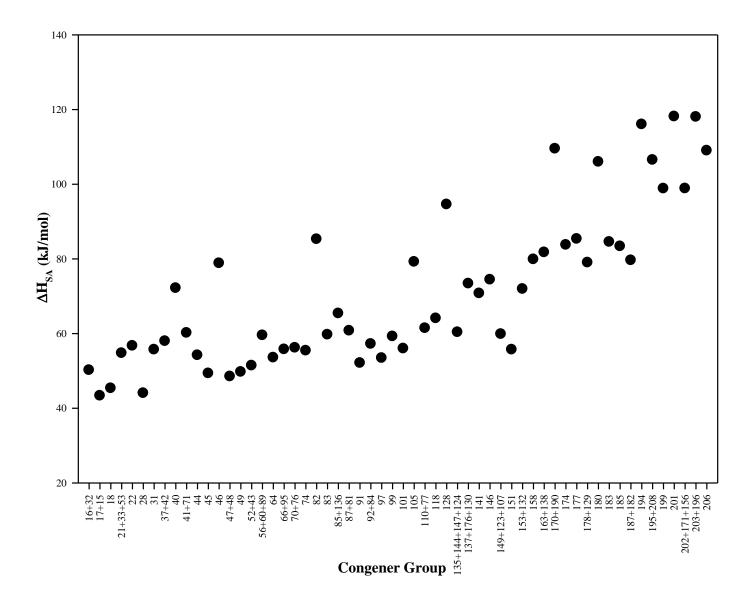


Figure 2.2.  $\Delta H_{SA}$  from this study plotted versus PCB congener number.

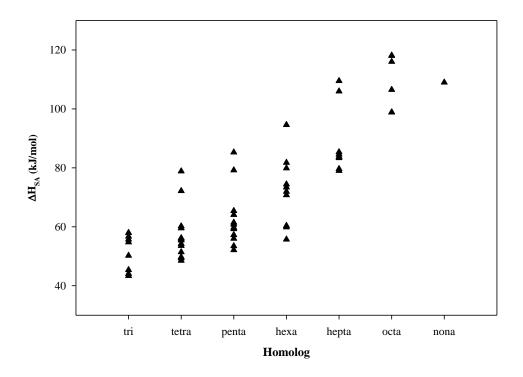


Figure 2.3.  $\Delta H_{SA}$  from this study plotted versus homolog.

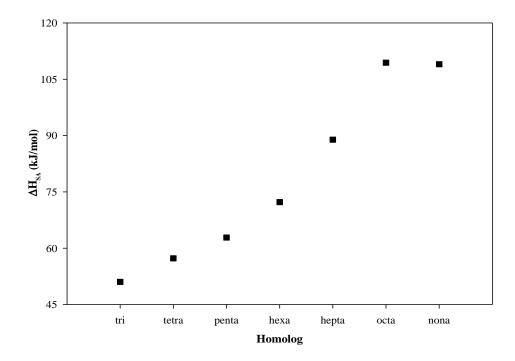


Figure 2.4.  $\Delta H_{SA}$  from this study averaged by homolog.

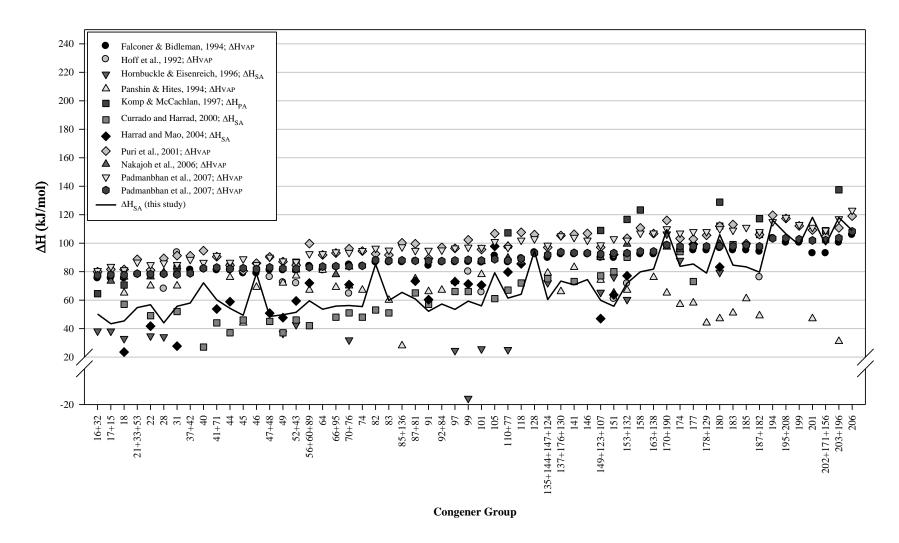
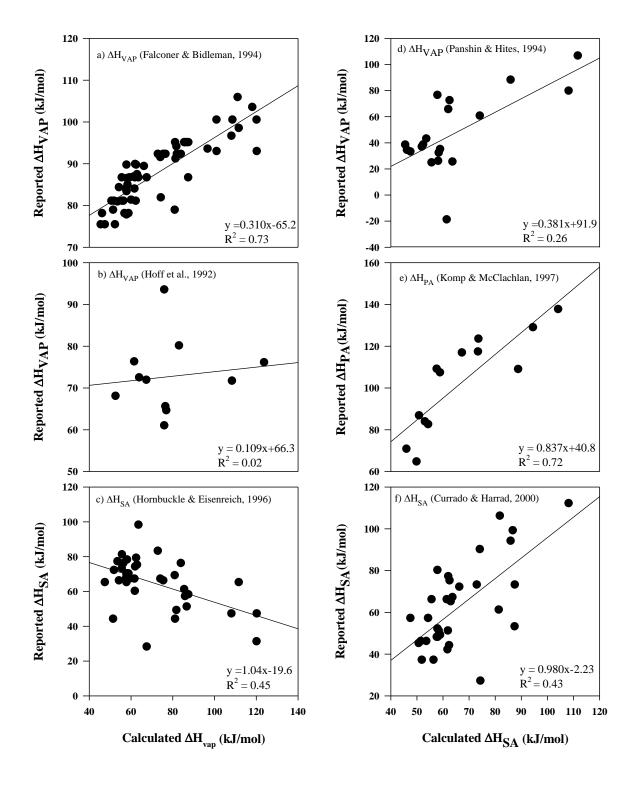


Figure 2.5. Comparison of calculated  $\Delta H_{SA}$  versus values reported in the literature.



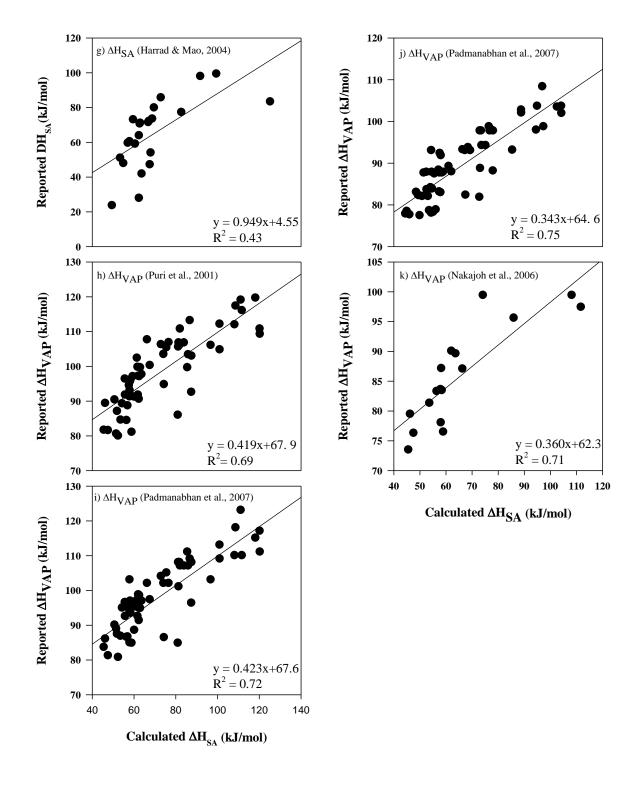


Figure 2.6. Literature reported  $\Delta H_{VAP}$ ,  $\Delta H_{SA}$ , and  $\Delta H_{PA}$  values versus  $\Delta H_{SA}$  calculated for Camden, NJ.

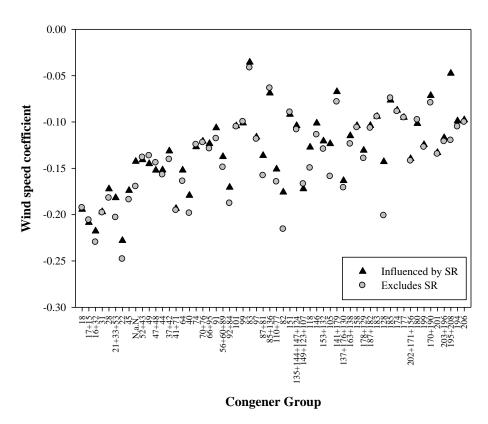


Figure 2.7. Wind speed coefficient plotted by PCB congener number.

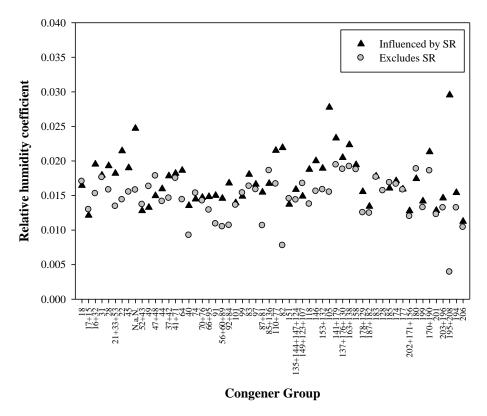


Figure 2.8. Relative humidity coefficients plotted by PCB congener number.

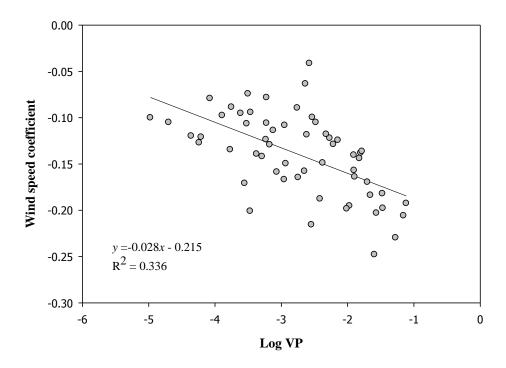


Figure 2.9. Relationship between wind speed coefficients and the log of vapor pressure of PCBs (p < 0.05).

## Chapter 3

# Source Apportionment of Polychlorinated Biphenyls in Chicago Air using Positive Matrix Factorization

#### **Abstract**

In order to effectively control the emissions of persistent organic pollutants, the sources responsible for the elevated urban concentrations must be identified. Recent studies have focused on identifying atmospheric PCB sources in urban areas, including Camden, New Jersey, but these studies have relied on data sets collected over a relatively short time. In this work, an advanced factor analysis method, Positive Matrix Factorization (PMF), was used to identify the dominant sources of polychlorinated biphenyls (PCBs) in the atmosphere of Chicago, IL using a data set collected over about 10 years (1996-2006) by the Integrated Atmospheric Deposition Network (IADN). PMF identified 6 factors which are assumed to represent source types in the Chicago area.

When compared to Aroclor compositions, only factor 1 (which comprised 23% of the PCB mass in the data set) was clearly identifiable as Aroclor 1242. Although PCB levels in most environmental compartments are declining gradually, with half lives on the order of 5 to 20 years, atmospheric PCB concentrations in Chicago displayed a marked increase starting around 2005. The PMF analysis suggests that this increase was due to factor 4 which appears to be a mixture of Aroclors 1248/1254 ( $R^2 = 0.674$ ), and factor 5 which resembles Aroclor 1254 ( $R^2 = 0.731$ ). Factors 4 and 5 comprise 24% and 16% of the total PCB mass in the data set. Factors 3 and 6 displayed an exponential decay with half lives of 3 and 5.8 years, respectively.

## Introduction

Despite the ban on PCB use and manufacture in the late 1970s, PCB levels are still elevated in urban areas, due to sources such as joint sealants, caulks, waste incineration, storage and disposal facilities, Superfund sites, and accidental releases [1-5]. Advective transport and deposition allow for transfer of PCBs from these urban zones to coastal regions where the chemicals can enter adjacent water bodies [6-10, 13]. This is currently a problem in the area surrounding Lake Michigan, where atmospheric PCB concentrations from the heavily industrialized Chicago, IL, region are impacting the concentrations within the lake [10-12]. In the Chicago region, atmospheric deposition is considered to be one of the largest sources of PCBs to Lake Michigan [9, 10].

The Integrated Atmospheric Deposition Network (IADN) was established in the early 1990s to characterize how PCBs and other contaminants originating in urban districts impact the nearby Great Lakes. The sampling performed by IADN has provided an extensive data set with over 10 years of measurements allowing the determination of long term trends. Generally, PCB concentrations measured in both the precipitation and gas phases in Chicago have decreased over the past 10 years [14], but concentrations still remain elevated compared to nonurban areas [9-12]. In this work, we investigate the hypothesis that the elevated levels of Chicago PCB concentrations are due to a construction boom that occurred in Chicago in the early part of the 2000s. We hypothesize that the older buildings that may have contained PCB laden materials were demolished, possibly releasing PCBs into the atmosphere.

PMF has been used successfully in many source apportionment studies, including studies designed to identify sources of PM2.5 [15, 16], volatile organics [17, 18], and semivolatile organic compounds such as polycyclic aromatic hydrocarbons (PAHs), [19, 20] in the atmosphere. PMF has also been used for congener fingerprinting of PCBs in sediment [21-23] and water [19, 24]. Recently, PMF was used for source identification of atmospheric PCBs in an attempt to isolate sources within the Camden/Philadelphia area [25, 26]. In this study, PMF was used to identify the types of sources that could be contributing to increased PCB concentrations measured in Chicago. The results will be compared to similar results reported for the PMF analysis using NJADN data from Camden, NJ. In addition, the factors identified through PMF will be examined for time trends, and the reasons behind the increase in PCB concentrations in the Chicago area post 2005 will be investigated.

# **Experimental Section:**

Data used in the PMF model was obtained from Ron Hites' Analytical Environmental Chemistry laboratory, Indiana University (IU) for the time period ranging from January 1996 through December 2006 (www.msc.ec.gc.ca/iadn/). Samples were collected as part of the Integrated Atmospheric Deposition Network (IADN), which was designed to characterize the loadings of persistent organic pollutants originating in the atmosphere into the urban and rural regions of the Great Lakes. The measurement of particle phase PCBs ceased in December 1996 and, therefore, the PMF model was run using only gas phase measurements. Full details regarding the collection and analysis of air samples can be found in the IADN Project Standard Operating, Sample Preparation,

and Gas Chromatography Procedures [27-30]. The procedures will also be summarized here.

# Sample Collection and Analysis:

The IADN monitoring equipment for the Chicago satellite location is set on the roof of the Farr Hall building situated on the Illinois Institute of Technology campus at an elevation of 200 meters. The Chicago IIT monitoring site is located 1.5 km west of Lake Michigan and is about 5.5 km south of downtown Chicago (Figure 3.1). The heaviest concentration of industrial facilities can be found approximately 10 to 20 km southeast of this sampling site in Chicago and northwest Indiana, as well as the occurrence of urban and industrial development being observed in all directions from the site [12, 14, 31, 32].

Air samples were collected over a 24 hour period once every 12 days using a modified Anderson High Volume air sampler (Hi-Vol) calibrated with a flow rate of 34 m<sup>3</sup> hr<sup>-1</sup>. Gas phase PCBs were captured using XAD-2 resin cartridges. The sampling medium was prepared at the IU and shipped along with supplies to the various sampling sites. All glassware used for sampling and analysis was washed with micro-90 soap and water, rinsed with hot tap water, air dried at room temperature overnight and placed in a muffle furnace for 6 hours at 450°C.

As part of the sampling medium preparation, Amberlite XAD-2 resin (20-60 mesh size, pore diameter 90A°) was rinsed with tap water followed by methanol and then underwent the following series of 24 hour soxhlet extractions: methanol, acetone, hexane, dichloromethane, hexane and a 50:50 mixture of acetone and hexane. The XAD-2 resin was then oven dried at 75°C for 8 hours and stored in an amber jar at -20°C until

cartridge assembly (or up to three months). The stainless steel cartridges were rinsed with hot tap water, Milli-Q DI water, allowed to dry overnight at room temperature, rinsed with dichloromethane and wrapped in aluminum foil until assembly with 40 grams of pre-cleaned, dry XAD-2 resin. New, pre-cleaned XAD-2 was utilized for the sample cartridges during the summer months (April through October), while recycled XAD-2 was used for the winter months (November through March).

Each fully assembled XAD-2 cartridge was wrapped in aluminum foil and placed in a solvent rinsed tin ointment can. The ointment can was sealed with a layer of Teflon tape followed by a layer of black electrical tape and stored at -20°C until shipment. After sample collection, the cartridges were returned to IU where the XAD-2 was transferred to a clean, glass jar capped with aluminum foil and stored at -20°C until analysis. Any samples that could not be transferred immediately were temporarily stored at 10°C until transfer could be completed.

The XAD-2 resin was extracted for 24 hours in a mixture of 50% acetone and 50% hexane using a soxhlet/condenser/round bottom flask apparatus. Prepared surrogate standards (PCBs 14, 65 and 166, PBDEs 77 and 166, carbon 13 labeled PBDE 209, Dibutyl chlorendate,  $\delta$ -HCH,  $d_{10}$  phenanthrene, and  $d_{10}$  pyrene) were injected onto the sample just before the extractions were started. Upon completion of the extraction, the round bottom flasks containing the solution of sample and solvent underwent rotary evaporation (Büchi Rotovapor Model R-114). The samples were concentrated to about 2 mL and transferred to hexane and separated into two fractions using deactivated silica chromatography. The silica was baked at 300°C overnight and deactivated using 3.5% deionized water by weight. PCBs were eluted within the first fraction using 25 mL of

hexane. The samples were then reduced to about 1 mL through another round of rotary evaporation, transferred with hexane to 4 mL vials and concentrated once more to 1 mL under a gentle stream of nitrogen gas. The PCB fraction was injected with internal standard (PCBs 30 and 204) and analyzed using an HP 6890 gas chromatograph equipped with 63Ni electron capture detector using a 60-m 0.25 mm i.d. DB-5 (5% diphenyl-dimethyl polysiloxane) capillary column with a film thickness of 0.1 μm.

IADN consists of a network of laboratories focused on characterizing the air quality in the Great Lakes region. To maintain a high level of consistency amongst laboratories quality assurance and control procedures are strictly followed. During the sampling period in question, laboratory blanks and matrix spikes represented 5% of the total number of collected field samples. Field blanks and field duplicates represented 10% of all field samples, until this number was reduced to 5% in July 2006. In addition, two new PCB standards were introduced in 2005 to further reduce variability between laboratories. The common calibration standard (CCS) and the calibration reference standard (CRS) replaced the Mullin 94 and Performance standards, respectively. The new standards are custom made by AccuStandard and consist of a suite of 84 individual PCB congeners most commonly found in air samples.

#### Positive Matrix Factorization Model:

PMF is an advanced factor analysis method that defines the sample matrix as a product of two unknown factor matrices with a residual matrix:

$$X = GF + E. (3.1)$$

The data matrix, X, is an  $(n \times m)$  matrix representing the observed data values where n is the sample number and m is the chemical species of interest. The G matrix  $(n \times p)$  characterizes the time variation of source (factor) strength, where n refers to the sample data and p refers to the number of sources or factors. F corresponds to the matrix of factor loadings which describes the chemical profiles of the sources or factors. E denotes the residual matrix or unexplained fraction remaining once G and F have been determined. The G and F matrices represent the PMF model solution and are determined by minimizing the sum of the squares, Q, via the following algorithm:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} (e_{ij} / s_{ij})^{2}$$
(3.2)

where  $e_{ij}$  represents the difference between the original data values, X, and the fitted values, G\*F, which are given by the model and  $s_{ij}$  is the uncertainty of the measured values in the data matrix. A more detailed description of the model can be found elsewhere [34].

## PMF Data Matrix:

PMF was performed on a data set containing 301 sampling events and 62 congeners (or co-eluting congener groups). Samples in which more than half of all congeners were below detection limit were discarded, and congeners that were below detection limit in more than 40% of samples were discarded. Also, congeners that were only measured during a portion of the 10-year study were discarded. As a result, all 62 of the congeners included in the modeling were on the list of "suite-PCBs" that are included in the new (post-2005) calibration standards. The suite-PCBs represent the list of

congeners and coeluting PCB GC peaks chosen by the IADN Steering Committee, and include congeners measured by both Canadian and United States agencies. The PMF model was originally run using the congener specific PCB data as reported by the IADN. When this was done, a marked increase in several factors was observed starting in January of 2005, when the new standards were adopted. A series of correction factors was provided with the raw data that allow the user to correct the post-2005 concentrations to reflect the pre-2005 standards. We employed this correction factor and found a much less pronounced change in the PMF results after January 2005. Thus we used the corrected data for all subsequent PMF analysis. A random number between 0 and one-half the detection limit was substituted for values below detection limit. There were two measured concentrations that appeared to be erroneous: PCB 33 on 11/19/05 and PCB 201 on 6/28/96. These concentrations were more than 200 times higher than the average for these congeners. For these two values, the geometric means of their concentrations were substituted in the data matrix.

# PMF Uncertainty Matrix:

PMF computes the error estimate  $(S_{ij})$  for each data point  $(X_{ij})$  based on the data point and its original error estimate. In the present study, the EM = -14 error model was used because it is recommended for general purpose environmental work:

$$S_{ij} = t_{ij} + u_{ij} \sqrt{\max(|x_{ij}|, |y_{ij}|)} + v_{ij} \max(|x_{ij}|, |y_{ij}|)$$
(3.3)

Where t is the congener- and sample-specific detection limit, u is the Poisson distribution (here designated as 0), v is the measurement precision, x is the observed data value, and y is the modeled value. The uncertainties associated with the measured PCB concentrations

include instrumental precision, extraction efficiency and sampling precision. The uncertainty matrix was estimated from the standard deviation of the surrogate recoveries, which were between 10 and 15%. We used a value of 15% uncertainty for all congeners, except below detection limit values and the two anomalous PCB values noted above, for which 3 times the normal uncertainty (45%) was used.

## **Results and Discussion:**

## Identification of Factors:

The PMF model was run requesting 2-9 factors and each run was initiated by varying the seed values starting with seed value 1 and ending with 10 for a total of 10 PMF runs. A critical step in PMF is determining the correct number of factors that provide clear, physically meaningful results and, at the same time, reduce matrix dimensionality. If errors are accurately measured the calculated Q value should be approximately equal to the theoretical Q value:

$$Q_{\text{theoretical}} = m \times n - p \times (m+n) \tag{3.4}$$

where *m* represents the number of samples, *n* refers to the number of PCB congeners used in the data matrix, and *p* is the number of factors requested [24, 25]. The calculated Q value is much higher than the theoretical Q value (Table 3.1). The calculated Q value depends very strongly on the uncertainty matrix, which can easily be in error. Thus a match between the theoretical and calculated Q values is not always a good indicator of the correct number of factors.

A second indicator of the correct number of factors is the point where the calculated Q values level off, which in this study occurs at about 6 factors. In addition,

the average relative standard deviation (RSD) of the 10 PMF runs can be used as a criterion for determining the correct number of factors. A marked increase in the RSD indicates that the PMF program cannot generate a stable solution using this number of factors. In the present case, the RSD increases considerably from 0.65% at 5 factors to 5.8% at 6 factors and again to 47.5% when 7 factors are requested. Our attempts to interpret the 5-factor model results revealed that when temperature trends were investigated, the Clausius-Clapeyron plots of all five factors displayed two distinct groups of points that were associated with the change in calibration standards that occurred after 2004. When the model was interpreted with 6 factors, the points on the Clausius-Clapeyron plots fell into only a single cluster for 5 of the six factors. All of the variability in the Clausius- Clapeyron plots had thus been sequestered to one factor (factor 2). This suggests that the model accounts for the change in analytical standards by collecting all of the associated variability and placing into one "junk" factor. To further support the decision that 6 is the appropriate number of factors, the fit between the measured and modeled PCB concentrations was examined. This comparison gives an R<sup>2</sup> of 0.981 for the sum of PCBs and was better than 0.8 for 49 of the 63 congeners. The 14 congeners that were not well described by the 6-factor model were PCBs 7+9, 19, 12, 13, 100, 70+76, 119, 81, 77, 167, 156, 199, 201, and 206.

## Resolved congener profiles:

The average molecular weights were 253, 278, 277, 314, 321 and 300 g/mol for factors 1 through 6, respectively. Factor 1, which contributes about 23% of the measured ΣPCBs in the Chicago area, is dominated by low molecular weight PCBs. Factors 2 and

3 are composed primarily of di-, tri-, tetra- and penta- chlorobiphenyls, and account for about 2% and 27% of  $\Sigma$ PCBs, respectively. Factors 4 and 5 consist predominantly of tetra- and penta- biphenyls, comprising about 24% and 16% of the measured  $\Sigma$ PCBs, respectively. Factor 6 represents 7% of the total PCB mass in the data set and consists of tri-, tetra-, penta-, hexa-, and hepta- PCB congeners (Figures 3.2 and 3.3).

# Identification of resolved factors:

When characterizing PCB PMF factors, is it assumed that if no substantial amount of environmental weathering or other alteration process has occurred, the congener composition of the resolved source profiles will match the composition of the Aroclor mixtures (Table 3.2) [24,25]. In order to identify the resolved source profiles, the congener patterns of the factors were compared to the congener patterns of Aroclor mixtures 1221, 1232, 1016, 1242, 1248, 1254, 1260, 1262 and 1268. The compositions of the Aroclor mixtures were taken from Rushneck et al. [35] and were measured using high resolution GC/MS analysis. To simulate the volatilization process, the reported congener concentrations were multiplied by their corresponding liquid vapor pressures [36]. Factor 1 was clearly identifiable as Aroclor 1242 ( $R^2 = 0.601$ ). Factors 2 and 6, which represent 2% and 7% of the total mass of PCBs, respectively, did not resemble any single Aroclor. Factors 3 and 4 resembled mixtures of Aroclors 1242/1248 ( $R^2 = 0.78$ ) and Aroclors 1248/1254 ( $R^2 = 0.674$ ), respectively. Factor 5 resembled only Aroclor 1254 ( $R^2 = 0.731$ ).

At present, only one other study used PMF to identify sources of PCBs in the atmosphere. Du et al. [25] utilized PMF to examine gas- and particle-phase PCB

concentrations in the atmosphere of Camden, NJ. For that study, the PMF model was applied to a much smaller data set of 74 sampling events and 52 PCB congeners or coeluting congener groups, as opposed to the data matrix for this study which included 301 sampling events and 62 PCB compounds. Du et al. resolved four factors in the atmosphere of the Camden, NJ whereas in this study six factors were generated.

Of the four factors reported for the Camden study (designated here as A, B, C, and D), the majority of the PCB mass was represented by factors A, B and C, while the total PCB mass in Chicago was dominated by factors 1, 3 and 4. Du et al. suggested that factor A was represented by a combination of low molecular weight Aroclors such as 1016, 1242 and 1248. In the current study, factor 1 strongly resembled only Aroclor 1242. In the atmosphere at Camden, NJ the source profile for factor B was most similar to the congener pattern of Aroclor 1254, as well as a mixture of Aroclors 1248 and 1254; and factors C and D did not bear any semblance to the Aroclors, though factor D was thought to resemble the average particle phase PCB concentration in Camden. In Chicago, factors 2 and 6 did not bear any resemblance to the Aroclors. Factors 3 and 4 were determined to be mixtures of Aroclors 1242/1248 and Aroclors 1248/1254, respectively, while factor 5 resembled only Aroclor 1254. The source profiles resolved for Camden, NJ more strongly resembled Aroclors than those of Chicago.

## Time and temperature dependence:

In order to determine if PCB concentrations in Chicago were declining, the relationship between the factor source concentrations (pg/m³) and date was examined (Figure 3.4). A gradual decline was seen for factors 3 and 6; however, factors 1, 2 and 5 remain fairly constant except for a noticeable increase occurring around July 1999. Factor 5 exhibits an increase after 2005. Factor 4 is increasing. The association between time and source was inspected more closely by plotting the factor source concentrations for January 2005 through December 2006. Factors 4 and 5 increase noticeably in August of 2005 with a second spike occurring in August of 2006, though this second increase is not as extreme for factor 4.

A multi-parameter linear regression of natural log of concentration ( $\ln C$ ) versus time (t) and inverse temperature (1/T in Kelvin) was performed to characterize any long term concentration trends that may be present in Chicago:

$$\ln C_{gas} = a_0 + a_1 \left(\frac{1}{T}\right) + a_2 t \tag{3.5}$$

The results confirm what is seen in the factor score versus time plots (Figure 3.4). Factors 3 and 6 are decreasing with time (p < 0.001), while factor 4 displays a statistically significant increase over time. Half lives and rate constants for factors 3 and 6 were computed from the slope values obtained through the multi-parameter linear regressions. Factors 3 and 6 are decreasing with half lives of 3.0 and 5.8 years, respectively, and their corresponding rate constants ( $\pm$  95% CL) are 0.228 $\pm$ 0.034 and 0.120 $\pm$ 0.040 per year.

Ambient PCB concentrations typically exhibit a correlation with temperature [11, 37, 38]. Application of the Clausius-Clapeyron equation demonstrates significant temperature dependence (p < 0.05) for five of the six factors (Table 3.3). Factors 1 and 3 show weak but significant relationships, while factors 4, 5 and 6 show stronger

relationships. When the 2005-2006 data is plotted separately from the 1996-2004 data, five of the six factors demonstrate significant temperature dependence (p<0.001), though for factor 2 this dependence is only evident for 2005-2006 (Tables 3.4, 3.5). Temperature dependence corresponding to the 2005-2006 time period is stronger than the 1996-2004 data for five of the six factors.

Declining PCB levels have been observed in sediment, soils, vegetation, water and biota [39-48]. Hillery et al. [37] noted that atmospheric PCB concentrations near Lake Superior have remained unchanged, though they did observe a reduction near Lakes Erie and Michigan. The environmental half lives of gas-phase PCBs were estimated to be about 6 years. Recently, Sun et al. [14] reported a decrease between 1997-2003 and 1996-2003 for PCBs measured in the precipitation and gas phase in Chicago, respectively. Half-lives equal to approximately 6-7 years were calculated for both phases. The half-lives calculated for factors 3 and 6 of this study are consistent with previously reported half lives for gas-phase PCBs measured in Chicago, suggesting that gas-phase PCB concentrations are declining. However, factor 6 (half life = 5.8 years) accounts for only 7% of the total PCB mass in the data set, so its decrease accounts for a relatively small fraction of the overall reduction in ΣPCBs. In contrast, factor 2 accounts for 27% of ΣPCBs so it accounts for a larger proportion of the overall decrease of ΣPCBs measured in Chicago.

The use of Aroclors for industrial purposes has been previously summarized (Table 3.6) [50]. The PMF model suggests that PCB levels explained by factor 1, which represent 23% of the total mass of PCBs in the dataset and resemble Aroclor 1242, are remaining constant. Aroclor 1242 was used mainly in transformers, hydraulic fluids and

plasticizers. Hsu et al. [1] used potential source contribution function (PSCF) modeling to locate PCB sources in Chicago. The study identified a transformer storage yard that spans about 10 acres and stores up to 500 transformers at a time. Due to the fact that the transformers stored at this site may have contained PCBs, either at the time of sampling or in the past, the storage yard could be considered a constant source of PCBs to the atmosphere in Chicago. However, the congener profiles for this site resembled a mixture of high and low MW congeners and thus a mixture of Aroclors [1]. An area of Chicago containing municipal sludge drying beds and the CID landfill was also named as a source of PCBs to the Chicago atmosphere by Hsu et al. Congener profiles at these sites were most similar to Aroclor 1242. Therefore, it is more likely that factor 1 can be explained by the volatilization of PCBs from the municipal sludge drying beds and the CID landfill.

Factor 2 which represents 2% of  $\Sigma$ PCBs in the dataset, does not resemble any of the Aroclor profiles and exhibits a weak but significant temperature dependence only for the 2005-2006 time period ( $R^2 = 0.356$ , p < 0.05). When the pre- and post-2005 datasets were examined separately from one another, it was found that the lower points were confined to the pre-2005 data set suggesting that factor 2 could be the PMF model's attempt to correct for the change in analytical standards that occurred in 2005. Factor 6, which also accounts for a relatively small fraction of the  $\Sigma$ PCBs (7%), also does not resemble any of the Aroclors but is declining with a half-life of 5.8 years. Unlike factor 2, factor 6 is strongly temperature dependent ( $R^2 = 0.631$ ). Factor 6 could represent the air/surface exchange of old emissions that are trapped in the soil or water column.

Factor 3, which represents 27% of the  $\Sigma$ PCBs in Chicago, resembles a mixture of Aroclor 1248 and Aroclor 1242. This factor is decreasing the fastest with a half-life of

3.0 years. Aroclors 1248 and 1242 were primarily used in transformers, hydraulics, and plasticizers. Sun et al. [14] attributed declining PCB concentrations to efforts aimed at reducing PCBs in the Great Lakes area resulting from the signing of the "Canada-United States Strategy for the Virtual Elimination of Persistent Toxic Substances in the Great Lakes Basin". The Strategy called for a 90% reduction, management and proper disposal of electrical equipment containing PCBs by the year 2006 [51]. It is possible that factor 3, which is decreasing the fastest, represents the successful removal and disposal of PCBs from the Chicago area.

Factor 4 resembles a combination of Aroclors 1248 and 1254, and represents 24% of the total mass of PCBs in the dataset. Unlike factor 3, factor 4 is steadily increasing with a doubling life of 8 years. Aroclors 1248 and 1254 were associated with hydraulics and plasticizers. Additional uses of Aroclor 1254 include inks, sealants, caulking compounds, dedusting agents, and pesticide extenders. A construction boom would most likely have involved the demolition of dozens of buildings over a period of 2 to 5 years. This would lead to the more steady increase in atmospheric PCBs over this period that would correspond to factor 4.

Factor 5, which accounts for 16% of the ΣPCBs in Chicago, resembles only
Aroclor 1254 and is also not decreasing over time. The data were more closely examined
for trends occurring within the pre- and post 2005 subdivisions (Tables 3.4, 3.5). Factor
5 appeared to have been decreasing with a half-life of 3.9 years until 2005 when it began
increasing with a doubling time of 1 year. This suggests the occurrence of a single event
is responsible for the sudden increase associated with factor 5. The increase could be
attributed to the change in laboratory standards. Because the resolved congener profile

strongly resembles Aroclor 1254, this increase could be due to the demolition of buildings that contained Aroclor 1254 in their building materials.

#### **Conclusions**

PMF was run using gas-phase PCB concentrations measured in Chicago during a 10 year period. Six factors were resolved. A steady increase over time was observed for factor 4, and a sudden increase was seen for factor 5. We speculate that these increases could be the result of a booming economy and the demolition of buildings originally built during the 1960s and 1970s, which could represent a fresh source of PCBs being released into the atmosphere. However, it should be noted that a new standard was introduced into the analytical process for IADN in 2005. It is possible that the increase seen in 2005 is an analytical artifact, related to this newer standard. Assuming this fresh source of PCBs is related to the redevelopment of Chicago, any PCBs released would most likely be dominated by Aroclor 1254, which was used in caulks and sealants [5]. The source resolved profiles generated by PMF supports this conclusion to some extent, as the profile for factor 5, which appears to be the main cause of the sudden rise in PCBs, most closely resembles the congener pattern of Aroclor 1254. However, this is weak evidence. The construction boom would most likely have involved the demolition of dozens of buildings over several years. This would lead to a long, low increase in atmospheric PCBs over this period, not the sudden jump observed in 2005. The increase in factor 5 observed in this data set is more indicative of a single dramatic event, perhaps the demolition of one very large building. For example, the Chicago Sun-Times building, located about 6 km south of the IIT sampling site, was demolished starting in October of

2004 [49]. This building was erected in 1955-1958

(http://www.glasssteelandstone.com/BuildingDetail/1067.php), during years when PCBs were being used in building materials. We offer this as an example of the kind of event that could be responsible for the PCB spike in 2005, but we have no evidence that the demolition of this building was the cause of the spike. Back trajectory analysis from days when factors 3 and 4 were particularly prevalent at IIT could indicate the direction of the source of this factor.

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Table 3.1. Descriptive parameters generated by the PMF model when various numbers of factors are requested.

Factors	Q <sub>CALC</sub>	Q <sub>THEOR</sub>	RSD
3	31100	17573	0.0035
4	26577	17210	0.0051
5	23929	16847	0.0065
6	21271	16484	0.0578
7	19570	16121	0.4749

Table 3.2. Source identification of factors through Aroclor composition.

	A1248	A1242	A1254	A1260	$\mathbb{R}^2$
F1	0%	105%	0%	0%	0.601
F2	56%	19%	22%	0%	0.559
F3	33%	59%	4%	0%	0.781
F4	39%	0%	62%	0%	0.674
F5	0%	7%	79%	0%	0.731
F6	42%	22%	8%	11%	0.217

Table 3.3. Results of the regressions of each factor vs. 1/T and time from 1996 through 2006 (Equation 3.5). N.S. = not signficant

Factor	$\mathbb{R}^2$	1/T (K <sup>-1</sup> )	$\mathbb{R}^2$	1/T (K <sup>-1</sup> )	t (yr <sup>-1</sup> )	$t_{1/2}$ (yr)
1	0.183	-3462±843	0.183	-3461±845	N.S.	
2	N.S.	N.S.	0.043	N.S.	$0.132 \pm 0.079$	
3	0.301	-5626±1047	0.568	-5723±825	$-0.228 \pm 0.034$	3.0
4	0.542	-5174±548	0.629	-5327±495	$0.084 \pm 0.020$	
5	0.494	-8852±1040	0.496	-8832± 1042	N.S.	
6	0.631	-12095±1091	0.672	-11950± 1031	$-0.120 \pm 0.040$	5.8

Table 3.4. Results of the regressions of each factor vs. 1/T and time from 1996 through 2004 (Equation 3.5). N.S. = not signficant

Factor	$\mathbb{R}^2$	1/T (K <sup>-1</sup> )	$\mathbb{R}^2$	1/T (K <sup>-1</sup> )	t (yr <sup>-1</sup> )	$t_{1/2}$ (yr)
1	0.152	$-3245 \pm 975$	0.154	$3232 \pm 976$	N.S.	
2	0.003	$-1031 \pm 2388$	N.S	N.S.	N.S.	
3	0.296	-5722 ± 1118	0.600	$-5431 \pm 845$	$-0.284 \pm 0.041$	2.4
4	0.575	$-5080 \pm 555$	0.592	$-5158 \pm 547$	$0.043 \pm 0.026$	
5	0.505	-9142 ± 1173	0.605	$-9059 \pm 1037$	$-0.177 \pm 0.047$	3.9
6	0.739	-11938 ± 917	0.751	- 11820 ± 898	$-0.069 \pm 0.041$	10.1

Table 3.5. Results of the regressions of each factor vs. 1/T and time from 2005 through 2006 (Equation 3.5). N.S. = not signficant

Factor	$\mathbb{R}^2$	1/T (K <sup>-1</sup> )	$\mathbb{R}^2$	1/T (K <sup>-1</sup> )	t (yr <sup>-1</sup> )	<i>t</i> <sub>1/2</sub> (yr)
1	0.430	$-4504 \pm 1474$	0.430	$-4505 \pm 1492$	N.S.	
2	0.356	$-4157 \pm 1606$	0.360	$-4169 \pm 1618$	N.S.	
3	0.416	$-6996 \pm 2519$	0.511	$-7206 \pm 2338$	$-0.710 \pm 0.496$	1.0
4	0.773	$-5896 \pm 907$	0.774	$-5883 \pm 915$	N.S.	
5	0.734	$-8499 \pm 1454$	0.836	$-8337 \pm 1155$	$0.679 \pm 0.247$	
6	0.455	$-13205 \pm 4440$	0.460	$-13382 \pm 4516$	N.S.	2.4

Table 3.6. Summary of Aroclor uses [50].

			Aroclor		
End use	1221	1232	1242	1248	1254
Capacitors	X				X
Transformers			X		X
Heat transfer			X		
Hydraulics/lubricants					
Hydraulic fluids		X	X	X	X
Vacuum pumps				X	X
Gas-transmission turbines	X		X		
Plasticizers					
Rubbers	X	X	X	X	X
Synthetic resins				X	X
Carbonless paper			X		
Miscellaneous					
Adhesives	X	X	X	X	X
Wax extenders			X		X
Dedusting agents					X
Inks					X
Cutting oils					X
Pesticide					X
Sealants and caulking compounds					X

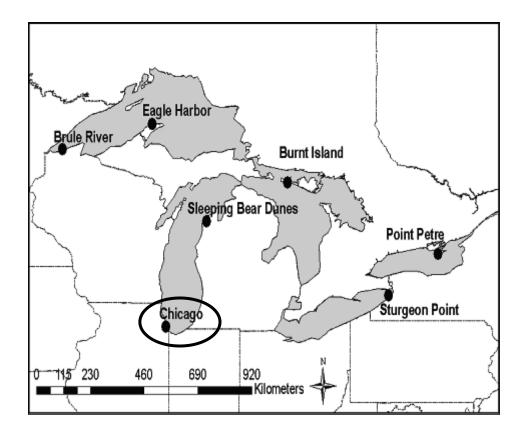


Figure 3.1. Location of IADN sampling sites [33].

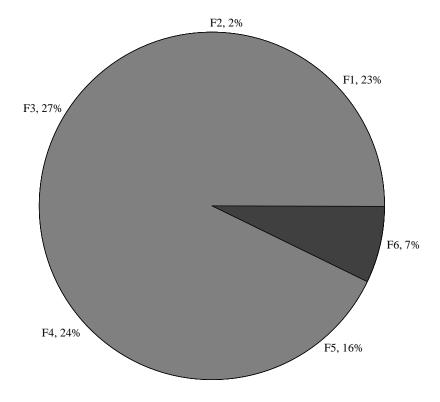


Figure 3.2. Contribution of factors to total PCB mass.

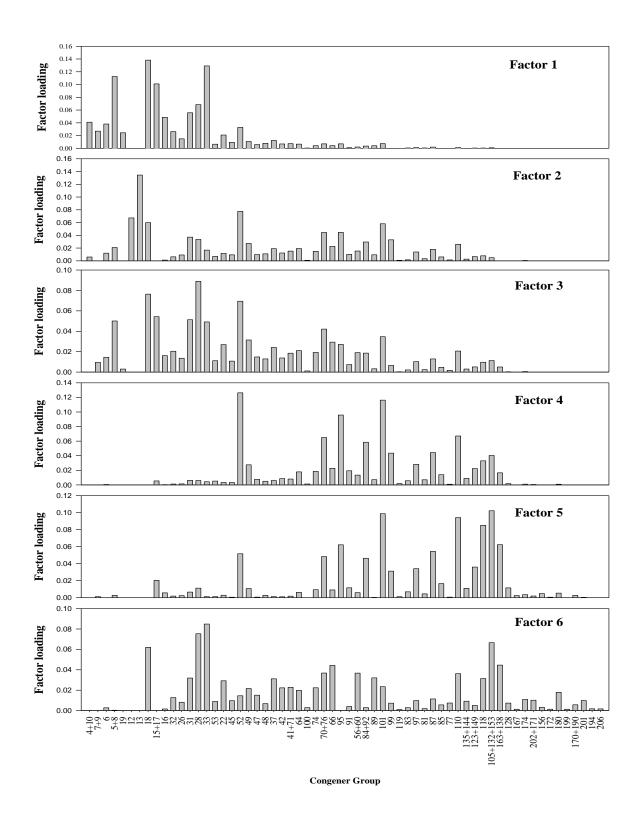


Figure 3.3. Normalized congener patterns of the six resolved source profiles. PCB congener numbers are plotted on the x-axis and fractional contribution to the sum of PCBs are plotted on the y-axis.

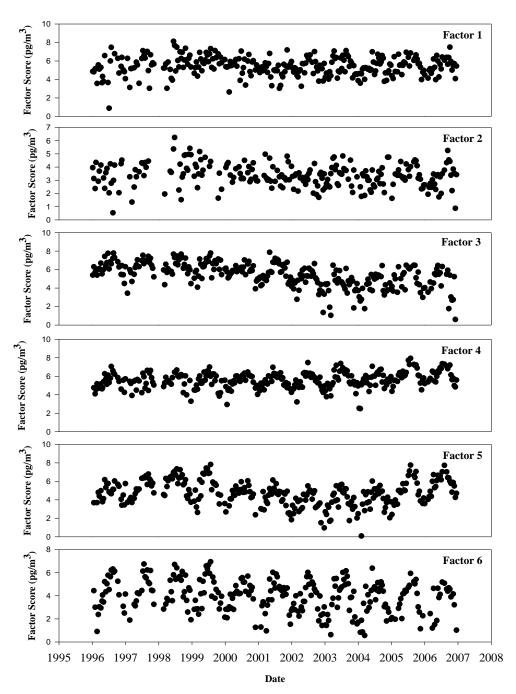


Figure 3.4. Factor score (pg/m³) versus time.

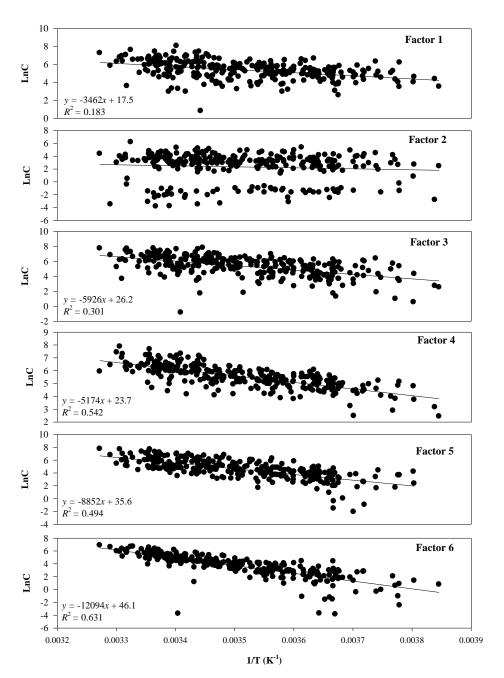


Figure 3.5. Temperature dependence of Factors 1 through 6.

## Chapter 4

## **Conclusions and future directions:**

The purpose of this thesis work was to better characterize atmospheric concentrations of PCBs in two urban locations. The persistence of PCBs has allowed for their transport within the atmosphere from the original source to other areas where they can be deposited onto surfaces. Atmospheric deposition is an important source of PCB contamination to many water bodies in the U.S., particularly in urban regions where elevated PCB concentrations are known to impact adjacent surface waters. The Delaware River and Lake Michigan are two systems affected by the PCB levels occurring in the surrounding atmosphere of Philadelphia, PA/Camden, NJ and Chicago, IL, respectively.

Meteorology can influence both contaminant concentrations and deposition. This can have an effect on atmospheric loadings of these contaminants to affected surfaces. It important to understand how meteorology and other atmospheric variables can influence gas-phase concentrations of persistent organic pollutants (POPs). In this work, the relationships between gas-phase PCBs in Camden, NJ and variables such as temperature, wind speed and direction, solar irradiance, relative humidity, and ozone were investigated using Statistical Analysis Software (SAS). For a three year period of data, strong relationships between wind speed, solar irradiance and relative humidity were seen when analyzed in the presence of temperature. The fact that the decrease in gas-phase PCB concentration with increasing wind speed is related to the vapor pressure of the PCB congener suggests that this effect may represent more than just dilution of gas-phase concentrations, since this physical process should affect all congeners equally. We

speculate that this effect may be related faster wind speeds driving faster uptake of gasphase PCBs into environmental compartments such as soil or vegetation.

While atmospheric PCB concentrations appear to be declining in most areas, an increase was observed for the Chicago area. Ten years of atmospheric PCB concentrations were made available by the Integrated Atmospheric Deposition Network (IADN) for analysis by Positive Matrix Factorization (PMF) in an attempt to identify the types of sources attributing to this increase. Factor 4, which accounts for 24% of the total PCB mass in the Chicago area during the time in question, represents the Aroclor 1254 signature. Prior to the ban of PCBs, Aroclor 1254 was used primarily in building materials such as caulks and fluorescent light ballasts. This work concludes that the increase of the Aroclor 1254 signature seen in Chicago could be related to the demolition of older buildings to make room for new construction in the area.

Overall, this thesis work provides new information for two unique urban atmospheres. However, this work raises some additional questions. We propose the following directions which may give rise to a better understanding of gas-phase PCB concentrations in the urban regions of Camden, NJ and Chicago, IL:

- 1. Expanding the time period utilized for statistical analysis in Camden, NJ. The acquisition of more recent gas-phase PCB concentrations and meteorological variables (O<sub>3</sub>, SR, WS, WD, and RH) could provide valuable information and allow for a more complete analysis of temporal trends.
- 2. The relationship observed between wind speed and vapor pressure of PCBs for Camden, NJ is an unusual phenomenon. The roles that soils and vegetation play in air-surface exchange of PCBs should be investigated further.

- 3. Some event or combination of events led to increasing PCB concentrations in Chicago, IL after 2005. The addition of more recent gas-phase PCB measurements into the PMF model could offer a clearer picture of source types present and temporal trends in the atmosphere surrounding Lake Michigan since 2005.
- 4. PMF has proven to be a successful tool in source apportionment of gas-phase PCBs. The model could be applied to PCB concentrations existing for other sampling sites situated on or near The Great Lakes, as well as PCB data available for other types of environmental media (i.e. aqueous phase, sediment core, etc.).

## Appendix I: Complete tables from Chapter 2.

Table I-1. Summary of gas phase linear regressions.

	<u>Ozone</u>	<u> </u>		Carbon	Monoxide	<u>:</u>	<u>NOx</u>			Wind S	<u>peed</u>		Inv Ten	<u>nperature</u>	
IUPAC	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-value	Slope
8+5	0.001	0.873	1.903	0.076	0.070	1.391	0.153	0.009	13.326	0.088	0.051	-0.214	0.011	0.494	-1.08E+03
18	0.003	0.724	4.045	0.072	0.078	1.303	0.113	0.026	11.021	0.216	0.002	-0.323	0.344	<.0001	-5.74E+03
17+15	0.005	0.670	4.571	0.089	0.055	1.350	0.128	0.020	10.771	0.262	0.001	-0.335	0.368	<.0001	-5.50E+03
16+32	0.021	0.351	11.047	0.040	0.196	1.003	0.078	0.066	9.539	0.249	0.001	-0.360	0.385	<.0001	-6.33E+03
31	0.032	0.242	13.710	0.025	0.310	0.784	0.048	0.154	7.399	0.234	0.001	-0.346	0.477	<.0001	-6.99E+03
28	0.032	0.243	11.388	0.026	0.299	0.668	0.054	0.131	6.520	0.263	0.000	-0.306	0.440	<.0001	-5.58E+03
21+33+53	0.042	0.250	13.549	0.003	0.775	0.280	0.011	0.568	4.134	0.245	0.003	-0.328	0.468	<.0001	-6.87E+03
22	0.054	0.131	19.813	0.014	0.448	0.661	0.034	0.232	7.006	0.228	0.001	-0.385	0.390	<.0001	-7.11E+03
45	0.043	0.177	13.974	0.022	0.342	0.651	0.036	0.216	5.695	0.247	0.001	-0.315	0.483	<.0001	-6.22E+03
46	0.080	0.191	22.133	0.020	0.519	-0.916	0.003	0.812	2.537	0.271	0.011	-0.423	0.560	<.0001	-9.77E+03
52+43	0.076	0.071	17.624	0.000	0.960	-0.033	0.003	0.737	1.484	0.197	0.003	-0.268	0.578	<.0001	-6.47E+03
49	0.046	0.163	13.588	0.006	0.609	0.331	0.016	0.418	3.536	0.208	0.002	-0.273	0.551	<.0001	-6.27E+03
47+48	0.024	0.319	10.148	0.018	0.392	0.574	0.029	0.265	5.034	0.208	0.002	-0.284	0.487	<.0001	-6.12E+03
44	0.083	0.058	19.427	0.000	0.969	0.027	0.006	0.616	2.332	0.212	0.002	-0.292	0.577	<.0001	-6.81E+03
37+42	0.057	0.119	17.625	0.007	0.590	0.406	0.019	0.371	4.532	0.167	0.006	-0.284	0.548	<.0001	-7.26E+03
41+71	0.054	0.130	18.354	0.004	0.676	0.338	0.018	0.382	4.754	0.221	0.001	-0.351	0.512	<.0001	-7.53E+03
64	0.092	0.045	20.923	0.000	0.895	0.093	0.009	0.542	2.888	0.213	0.002	-0.299	0.542	<.0001	-6.73E+03
40	0.165	0.019	32.417	0.011	0.559	-0.694	0.010	0.577	-4.887	0.161	0.021	-0.323	0.540	<.0001	-8.97E+03
74	0.089	0.050	20.026	0.000	0.993	0.006	0.003	0.717	1.679	0.178	0.004	-0.267	0.606	<.0001	-6.95E+03
70+76	0.114	0.025	22.558	0.003	0.715	-0.248	0.000	0.978	0.126	0.174	0.005	-0.262	0.633	<.0001	-7.05E+03
66+95	0.132	0.015	23.951	0.007	0.595	-0.357	0.001	0.888	-0.639	0.180	0.004	-0.263	0.639	<.0001	-7.00E+03
91	0.139	0.013	22.496	0.003	0.709	-0.230	0.000	0.900	-0.522	0.178	0.004	-0.240	0.668	<.0001	-6.56E+03
56+60+89	0.161	0.007	29.057	0.012	0.483	-0.518	0.000	0.908	-0.579	0.168	0.006	-0.280	0.598	<.0001	-7.45E+03
92+84	0.133	0.015	26.713	0.004	0.677	-0.311	0.000	0.970	0.190	0.208	0.002	-0.314	0.544	<.0001	-7.17E+03
101	0.145	0.011	24.821	0.015	0.431	-0.523	0.006	0.621	-2.222	0.157	0.008	-0.243	0.656	<.0001	-7.02E+03
99	0.121	0.021	23.999	0.005	0.637	-0.334	0.002	0.779	-1.337	0.147	0.010	-0.249	0.652	<.0001	-7.41E+03
83	0.121	0.004	30.035	0.003	0.194	-1.162	0.002	0.081	-11.513	0.094	0.045	-0.205	0.683	<.0001	-7.47E+03
97	0.156	0.004	26.624	0.041	0.134	-0.436	0.072	0.700	-1.793	0.167	0.043	-0.260	0.564	<.0001	-6.72E+03
71	0.130	0.006	20.024	0.010	0.520	-0.430	0.004	0.700	-1./93	0.107	0.000	-0.200	0.504	<.0001	-0.72E+03

	Ozone	1 -	Las		Monoxide	-	NOx	î	l an	Wind S		l an		<u>nperature</u>	
IUPAC	R <sup>2</sup>	p-value	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-value	Slope
87+81	0.127	0.042	22.847	0.047	0.225	-1.145	0.006	0.676	-2.959	0.184	0.013	-0.277	0.602	<.0001	-7.60E+03
85+136	0.087	0.052	24.354	0.000	0.995	0.005	0.001	0.841	-1.140	0.092	0.045	-0.236	0.554	<.0001	-8.15E+03
110+77	0.171	0.005	33.002	0.010	0.524	-0.518	0.002	0.788	-1.485	0.181	0.004	-0.320	0.523	<.0001	-7.68E+03
82	0.168	0.006	44.877	0.016	0.417	-0.905	0.002	0.758	-2.324	0.133	0.015	-0.375	0.525	<.0001	-1.05E+04
151	0.140	0.012	23.929	0.012	0.476	-0.464	0.006	0.608	-2.257	0.147	0.010	-0.231	0.677	<.0001	-6.99E+03
135+144+147+124	0.156	0.008	27.453	0.019	0.375	-0.626	0.009	0.534	-2.973	0.153	0.009	-0.256	0.669	<.0001	-7.55E+03
149+123+107	0.150	0.010	30.176	0.009	0.553	-0.472	0.003	0.748	-1.731	0.190	0.003	-0.320	0.522	<.0001	-7.49E+03
118	0.127	0.042	23.773	0.066	0.150	-1.407	0.011	0.564	-4.238	0.183	0.013	-0.288	0.615	<.0001	-7.99E+03
146	0.186	0.004	34.843	0.019	0.373	-0.731	0.006	0.626	-2.714	0.143	0.011	-0.288	0.741	<.0001	-9.24E+03
153+132	0.179	0.004	33.830	0.023	0.330	-0.791	0.008	0.564	-3.179	0.158	0.008	-0.300	0.707	<.0001	-8.94E+03
105	0.146	0.011	36.469	0.015	0.431	-0.749	0.000	0.993	0.057	0.145	0.012	-0.332	0.621	<.0001	-9.81E+03
141	0.142	0.012	30.562	0.023	0.331	-0.802	0.014	0.452	-4.201	0.117	0.023	-0.261	0.666	<.0001	-8.80E+03
137+176+130	0.087	0.097	28.039	0.061	0.165	-1.943	0.002	0.821	-2.376	0.128	0.041	-0.343	0.391	<.0001	-9.11E+03
163+138	0.165	0.006	36.451	0.023	0.327	-0.894	0.010	0.527	-3.909	0.145	0.011	-0.322	0.720	<.0001	-1.01E+04
158	0.157	0.008	34.568	0.017	0.398	-0.748	0.014	0.450	-4.524	0.134	0.015	-0.300	0.730	<.0001	-9.89E+03
178+129	0.109	0.031	30.361	0.020	0.371	-0.820	0.002	0.797	-1.610	0.129	0.018	-0.302	0.663	<.0001	-9.79E+03
187+182	0.122	0.022	27.249	0.014	0.444	-0.617	0.005	0.640	-2.547	0.116	0.026	-0.252	0.829	<.0001	-9.86E+03
183	0.140	0.012	33.174	0.010	0.521	-0.579	0.003	0.711	-2.265	0.123	0.020	-0.292	0.788	<.0001	-1.05E+04
128	0.171	0.006	48.291	0.018	0.390	-1.000	0.000	0.919	-0.807	0.138	0.014	-0.396	0.584	<.0001	-1.17E+04
185	0.139	0.013	32.051	0.008	0.574	-0.492	0.003	0.721	-2.119	0.110	0.028	-0.268	0.815	<.0001	-1.03E+04
174	0.155	0.008	34.846	0.017	0.399	-0.758	0.006	0.615	-3.061	0.115	0.024	-0.282	0.779	<.0001	-1.04E+04
177	0.160	0.008	34.715	0.022	0.338	-0.857	0.009	0.536	-3.748	0.100	0.039	-0.260	0.768	<.0001	-1.06E+04
202+171+156	0.151	0.009	38.915	0.005	0.653	-0.460	0.002	0.759	-2.118	0.130	0.016	-0.341	0.836	<.0001	-1.22E+04
180	0.142	0.012	40.846	0.011	0.501	-0.743	0.004	0.683	-3.051	0.107	0.030	-0.334	0.817	<.0001	-1.30E+04
199	0.122	0.020	34.401	0.004	0.681	-0.414	0.002	0.766	-2.022	0.127	0.018	-0.331	0.863	<.0001	-1.22E+04
170+190	0.198	0.002	49.707	0.032	0.245	-1.312	0.018	0.389	-6.605	0.093	0.044	-0.321	0.824	<.0001	-1.35E+04
201	0.143	0.011	43.965	0.007	0.582	-0.652	0.007	0.587	-4.345	0.111	0.027	-0.365	0.881	<.0001	-1.45E+04
203+196	0.147	0.010	44.291	0.009	0.541	-0.720	0.008	0.555	-4.690	0.107	0.030	-0.356	0.891	<.0001	-1.45E+04
195+208	0.220	0.001	60.095	0.049	0.148	-1.871	0.033	0.237	-10.365	0.063	0.101	-0.303	0.591	<.0001	-1.31E+04

	<u>Ozone</u>			Carbon	Monoxide	<u>!</u>	<u>NOx</u>			Wind S <sub>1</sub>	<u>peed</u>		Inv Ten	<u>iperature</u>	
IUPAC	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-	Slope	$\mathbb{R}^2$	p-value	Slope
194	0.174	0.005	47.847	0.035	0.225	-1.409	0.028	0.278	-8.512	0.097	0.039	-0.337	0.874	<.0001	-1.42E+04
206	0.184	0.004	47.417	0.041	0.194	-1.474	0.057	0.123	-11.765	0.069	0.089	-0.275	0.760	<.0001	-1.34E+04
∑PCBs (incl. 8+5)	0.076	0.071	18.263	0.003	0.715	0.247	0.017	0.402	3.821	0.225	0.001	-0.297	0.572	<.0001	-6.68E+03
<b>∑PCBs (w/o 8+5)</b>	0.088	0.050	20.209	0.001	0.870	0.114	0.009	0.548	2.812	0.215	0.002	-0.297	0.601	<.0001	-7.00E+03

Table I-1. Summary of gas phase linear regressions (continued).

	Sine Wi	nd Directio	on	Cosine '	Wind Direc	tion	Station	Pressure		Relative	Humidity		Solar R	adiation	
IUPAC	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
8+5	0.014	0.453	0.182	0.019	0.375	-0.179	0.005	0.666	-0.009	0.007	0.602	0.005	0.043	0.176	1.847
18	0.002	0.771	-0.068	0.025	0.302	-0.200	0.030	0.258	-0.023	0.174	0.005	0.025	0.017	0.394	1.124
17+15	0.000	0.902	-0.027	0.034	0.241	-0.214	0.032	0.259	-0.022	0.176	0.006	0.023	0.023	0.337	1.203
16+32	0.001	0.820	-0.056	0.023	0.331	-0.196	0.039	0.200	-0.027	0.154	0.009	0.024	0.045	0.168	1.881
31	0.005	0.652	-0.109	0.020	0.365	-0.181	0.029	0.269	-0.023	0.180	0.004	0.026	0.039	0.202	1.730
28	0.004	0.670	-0.086	0.035	0.222	-0.203	0.034	0.230	-0.021	0.206	0.002	0.023	0.039	0.197	1.455
21+33+53	0.003	0.776	-0.074	0.034	0.308	-0.207	0.008	0.624	-0.010	0.201	0.009	0.023	0.049	0.217	1.694
22	0.001	0.891	-0.037	0.019	0.377	-0.199	0.046	0.160	-0.033	0.123	0.020	0.024	0.065	0.095	2.525
45	0.002	0.785	-0.058	0.030	0.264	-0.197	0.051	0.140	-0.028	0.183	0.004	0.023	0.052	0.135	1.784
46	0.000	0.961	0.021	0.005	0.740	-0.105	0.000	0.988	0.001	0.239	0.018	0.031	0.056	0.277	2.365
52+43	0.012	0.482	-0.143	0.014	0.438	-0.131	0.032	0.247	-0.021	0.155	0.008	0.020	0.057	0.120	1.766
49	0.012	0.473	-0.144	0.020	0.360	-0.153	0.037	0.212	-0.022	0.198	0.002	0.023	0.031	0.255	1.290
47+48	0.011	0.499	-0.141	0.032	0.248	-0.200	0.037	0.213	-0.023	0.211	0.002	0.024	0.021	0.351	1.101
44	0.007	0.583	-0.117	0.016	0.420	-0.143	0.039	0.198	-0.024	0.156	0.008	0.021	0.070	0.082	2.071
37+42	0.000	0.968	0.009	0.016	0.410	-0.160	0.028	0.276	-0.022	0.133	0.015	0.022	0.076	0.070	2.359
41+71	0.005	0.650	-0.114	0.016	0.417	-0.169	0.038	0.208	-0.028	0.168	0.006	0.026	0.049	0.150	2.026
64	0.004	0.685	-0.089	0.020	0.355	-0.167	0.048	0.152	-0.027	0.155	0.008	0.022	0.075	0.072	2.186
40	0.007	0.653	-0.142	0.021	0.417	-0.201	0.028	0.350	-0.024	0.103	0.068	0.020	0.092	0.086	2.825
74	0.012	0.477	-0.151	0.019	0.378	-0.156	0.041	0.188	-0.025	0.163	0.007	0.022	0.058	0.117	1.866
70+76	0.012	0.487	-0.147	0.015	0.426	-0.140	0.046	0.163	-0.026	0.149	0.010	0.021	0.074	0.074	2.102

	Sine Wi	nd Directio	n	Cosine '	Wind Direc	tion	Station	Pressure		Relative	Humidity		Solar R	adiation	
IUPAC	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
66+95	0.011	0.493	-0.143	0.014	0.439	-0.134	0.044	0.175	-0.025	0.136	0.014	0.020	0.090	0.047	2.293
91	0.011	0.501	-0.129	0.033	0.235	-0.188	0.055	0.124	-0.026	0.123	0.020	0.017	0.120	0.021	2.418
56+60+89	0.005	0.663	-0.100	0.012	0.486	-0.133	0.044	0.170	-0.028	0.094	0.043	0.018	0.116	0.024	2.857
92+84	0.005	0.642	-0.108	0.010	0.521	-0.124	0.054	0.130	-0.031	0.104	0.033	0.019	0.112	0.027	2.833
101	0.019	0.369	-0.185	0.017	0.407	-0.142	0.043	0.179	-0.024	0.140	0.012	0.020	0.081	0.062	2.142
99	0.028	0.280	-0.235	0.030	0.261	-0.204	0.044	0.171	-0.026	0.148	0.010	0.021	0.070	0.082	2.123
83	0.023	0.335	-0.210	0.024	0.322	-0.181	0.074	0.078	-0.033	0.144	0.012	0.021	0.087	0.055	2.346
97	0.028	0.274	-0.232	0.008	0.568	-0.102	0.039	0.200	-0.024	0.165	0.006	0.022	0.055	0.126	1.828
87+81	0.009	0.605	-0.131	0.031	0.328	-0.194	0.022	0.411	-0.017	0.151	0.026	0.020	0.089	0.092	2.231
85+136	0.009	0.547	-0.157	0.026	0.300	-0.225	0.047	0.158	-0.032	0.131	0.016	0.024	0.049	0.147	2.120
110+77	0.026	0.300	-0.261	0.004	0.674	-0.089	0.041	0.187	-0.029	0.146	0.011	0.025	0.072	0.008	2.487
82	0.000	0.976	-0.010	0.012	0.483	-0.202	0.066	0.092	-0.051	0.044	0.173	0.018	0.172	0.005	5.253
151	0.030	0.262	-0.226	0.021	0.347	-0.158	0.056	0.123	-0.027	0.153	0.009	0.020	0.071	0.080	1.973
135+144+147+124	0.023	0.322	-0.217	0.025	0.310	-0.185	0.057	0.118	-0.030	0.137	0.013	0.021	0.090	0.048	2.412
149+123+107	0.054	0.130	-0.370	0.018	0.390	-0.176	0.037	0.211	-0.027	0.153	0.009	0.025	0.044	0.170	1.903
118	0.011	0.555	-0.155	0.037	0.287	-0.219	0.030	0.337	-0.020	0.185	0.012	0.023	0.075	0.124	2.123
146	0.037	0.209	-0.319	0.022	0.334	-0.205	0.074	0.074	-0.040	0.124	0.019	0.023	0.126	0.018	3.317
153+132	0.032	0.253	-0.304	0.017	0.399	-0.180	0.069	0.085	-0.038	0.132	0.015	0.023	0.108	0.029	3.048
105	0.005	0.652	-0.135	0.006	0.615	-0.125	0.083	0.060	-0.049	0.120	0.023	0.026	0.137	0.015	4.009
141	0.357	0.357	-0.236	0.039	0.202	-0.271	0.075	0.071	-0.040	0.153	0.009	0.026	0.087	0.053	2.764
137+176+130	0.015	0.491	-0.259	0.000	0.935	-0.024	0.031	0.330	-0.029	0.148	0.027	0.029	0.028	0.352	1.860
163+138	0.034	0.231	-0.339	0.026	0.296	-0.246	0.073	0.077	-0.044	0.141	0.012	0.027	0.103	0.034	3.334
158	0.046	0.163	-0.381	0.038	0.204	-0.290	0.101	0.036	-0.050	0.139	0.013	0.026	0.094	0.043	3.098
178+129	0.050	0.149	-0.414	0.011	0.495	-0.163	0.077	0.072	-0.045	0.096	0.043	0.022	0.118	0.024	3.593
187+182	0.011	0.507	-0.167	0.012	0.481	-0.147	0.040	0.196	-0.030	0.078	0.071	0.018	0.146	0.011	3.462
183	0.036	0.218	-0.343	0.023	0.322	-0.231	0.082	0.060	-0.046	0.123	0.092	0.025	0.111	0.027	3.424
128	0.003	0.733	-0.126	0.009	0.557	-0.178	0.077	0.071	-0.058	0.096	0.043	0.029	0.166	0.007	5.401
185	0.029	0.272	-0.297	0.020	0.359	-0.207	0.063	0.101	-0.039	0.117	0.023	0.024	0.115	0.025	3.369
174	0.034	0.229	-0.335	0.022	0.338	-0.223	0.069	0.085	-0.042	0.112	0.026	0.024	0.116	0.023	3.491
177	0.026	0.304	-0.286	0.014	0.457	-0.173	0.035	0.232	-0.031	0.088	0.053	0.021	0.116	0.025	3.433

	Sine Wi	nd Directio	n	Cosine V	Wind Direc	tion	Station	Pressure		Relative	Humidity		Solar Ra	adiation	
IUPAC	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
202+171+156	0.025	0.305	-0.324	0.007	0.595	-0.141	0.076	0.071	-0.050	0.071	0.081	0.021	0.167	0.006	4.747
180	0.047	0.158	-0.480	0.026	0.292	-0.300	0.087	0.052	-0.058	0.099	0.037	0.028	0.124	0.019	4.433
199	0.019	0.373	-0.278	0.019	0.369	-0.233	0.087	0.051	-0.053	0.079	0.065	0.022	0.166	0.006	4.657
170+190	0.044	0.172	-0.478	0.027	0.288	-0.311	0.081	0.061	-0.058	0.090	0.048	0.027	0.148	0.010	4.971
201	0.025	0.306	-0.375	0.019	0.378	-0.269	0.095	0.042	-0.065	0.059	0.113	0.023	0.187	0.003	5.812
203+196	0.027	0.286	-0.389	0.019	0.369	-0.273	0.096	0.040	-0.065	0.062	0.102	0.023	0.188	0.003	5.796
195+208	0.000	0.961	0.020	0.002	0.753	-0.107	0.070	0.084	-0.061	0.017	0.404	0.013	0.268	0.000	7.682
194	0.020	0.361	-0.331	0.030	0.263	-0.336	0.059	0.112	-0.050	0.061	0.108	0.023	0.187	0.003	5.746
206	0.003	0.739	-0.119	0.011	0.505	-0.198	0.032	0.252	-0.038	0.033	0.244	0.016	0.167	0.007	5.240
∑PCBs (incl. 8+5)	0.009	0.542	-0.128	0.020	0.365	-0.158	0.047	0.156	-0.026	0.171	0.005	0.022	0.064	0.098	1.946
<b>∑PCBs</b> (w/o 8+5)	0.012	0.483	-0.151	0.021	0.350	-0.167	0.049	0.148	-0.027	0.165	0.006	0.022	0.071	0.081	2.097

Table I-2. Summary of particle phase linear regressions (vapor pressures taken from Falconer and Bidleman, 1994).

		Ozone			Carbon	Monoxide		NOx			Wind S	peed		Inv Ten	nperature	
CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	R <sup>2</sup>	p-value	Slope	WS P <sup>2</sup>	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
8+5	-0.808	0.004	0.743	-3.841	0.013	0.523	0.690	0.000	0.956	-0.488	0.002	0.789	0.037	0.000	0.972	-1.47E-
18	-1.115	0.038	0.195	-14.884	0.043	0.169	1.183	0.048	0.142	8.896	0.001	0.813	-0.030	0.006	0.610	-1.94E-
17+15	-1.155	0.011	0.612	-7.257	0.001	0.855	0.249	0.004	0.757	-3.470	0.002	0.813	0.042	0.008	0.675	-2.72E-
16+32	-1.275	0.019	0.366	-10.376	0.032	0.237	1.012	0.024	0.310	6.164	0.006	0.626	-0.061	0.011	0.496	-2.57E-
31	-1.467	0.044	0.163	-12.606	0.061	0.098	1.115	0.065	0.087	8.134	0.014	0.443	-0.076	0.003	0.710	-1.11E-
28	-1.474	0.047	0.149	-13.295	0.099	0.033	1.450	0.079	0.059	9.115	0.015	0.425	-0.080	0.006	0.601	-1.60E-
21+33+53	-1.564	0.022	0.472	-9.286	0.026	0.433	0.963	0.005	0.745	3.297	0.003	0.784	0.044	0.012	0.597	-3.10E-
22	-1.594	0.083	0.052	-20.427	0.068	0.080	1.384	0.061	0.098	9.238	0.013	0.444	-0.089	0.002	0.789	9.45E+
45	-1.655	0.017	0.491	-8.935	0.001	0.856	0.220	0.000	0.992	0.107	0.000	0.950	0.010	0.003	0.773	1.38E+
46	-1.700	0.062	0.219	-16.844	0.038	0.341	1.254	0.000	0.960	0.555	0.007	0.695	0.068	0.000	0.985	1.22E+
52+43	-1.801	0.076	0.063	-17.651	0.048	0.144	1.048	0.047	0.150	7.301	0.000	0.906	-0.012	0.000	0.899	-4.04E-
49	-1.781	0.059	0.103	-15.026	0.073	0.069	1.251	0.061	0.100	8.028	0.002	0.760	-0.031	0.007	0.595	-1.64E-
47+48	-1.821	0.042	0.173	-13.297	0.042	0.172	0.997	0.036	0.207	6.517	0.009	0.543	-0.065	0.001	0.830	6.95E+

		Ozone			Carbon	Monoxide		NOx			Wind S	peed		Inv Ten	nperature	
CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\frac{WS}{D^2}$	p-value	Slope	$\mathbb{R}^2$	p-value	Slo
44	-1.901	0.047	0.149	-13.556	0.065	0.087	1.199	0.043	0.165	6.910	0.006	0.606	-0.053	0.000	0.938	-2
37+42	-1.905	0.045	0.158	-16.212	0.100	0.033	1.812	0.057	0.110	9.672	0.008	0.567	-0.072	0.000	0.921	3.
41+71	-1.971	0.029	0.260	-12.901	0.045	0.156	1.212	0.044	0.164	8.395	0.005	0.636	-0.059	0.000	0.952	2
64	-1.891	0.038	0.193	-12.824	0.079	0.059	1.381	0.057	0.109	8.303	0.013	0.460	-0.079	0.000	0.927	-3
40	-2.012	0.064	0.214	-24.429	0.045	0.301	1.951	0.041	0.319	15.456	0.063	0.217	-0.303	0.065	0.208	1
74	-2.145	0.064	0.091	-19.245	0.054	0.122	1.322	0.037	0.199	7.773	0.013	0.456	-0.093	0.002	0.793	ç
70+76	-2.269	0.038	0.195	-14.055	0.034	0.219	0.998	0.033	0.230	6.880	0.003	0.708	-0.044	0.003	0.705	1
66+95	-2.214	0.064	0.089	-16.563	0.048	0.143	1.075	0.045	0.158	7.308	0.003	0.721	-0.038	0.005	0.642	1
91	-2.322	0.119	0.030	-26.367	0.090	0.060	1.704	0.050	0.167	8.932	0.000	0.948	0.009	0.002	0.787	
56+60+89	-2.375	0.025	0.298	-11.501	0.026	0.286	0.883	0.030	0.251	6.689	0.006	0.600	-0.063	0.005	0.631	
92+84	-2.414	0.046	0.153	-14.448	0.054	0.122	1.169	0.043	0.165	7.422	0.002	0.761	-0.034	0.005	0.645	
101	-2.478	0.067	0.084	-16.448	0.028	0.270	0.793	0.041	0.178	6.811	0.001	0.804	-0.026	0.009	0.543	
99	-2.531	0.074	0.067	-19.318	0.055	0.117	1.245	0.036	0.208	7.095	0.000	0.968	-0.005	0.001	0.842	
83	-2.574	0.096	0.049	-22.502	0.049	0.166	1.221	0.026	0.316	6.220	0.001	0.892	-0.018	0.000	0.972	
97	-2.614	0.030	0.252	-11.184	0.023	0.313	0.739	0.044	0.162	7.188	0.003	0.734	-0.036	0.012	0.465	
87+81	-2.649	0.055	0.249	-17.150	0.066	0.207	1.786	0.040	0.328	11.460	0.003	0.810	0.045	0.000	0.987	
85+136	-2.634	0.014	0.433	-8.347	0.047	0.149	1.140	0.010	0.516	3.658	0.002	0.797	0.030	0.002	0.753	
110+77	-2.744	0.015	0.421	-8.619	0.012	0.467	0.584	0.031	0.243	6.577	0.009	0.526	-0.074	0.022	0.321	
82	-2.544	0.031	0.240	-13.893	0.077	0.062	1.630	0.041	0.176	8.420	0.018	0.376	-0.114	0.024	0.309	
151	-2.758	0.091	0.042	-18.523	0.051	0.132	1.037	0.065	0.087	8.304	0.000	0.906	-0.012	0.006	0.607	
135+144+147+124	-2.948	0.135	0.012	-27.129	0.061	0.098	1.366	0.062	0.095	9.735	0.002	0.762	-0.037	0.025	0.291	
149+123+107	-2.955	0.065	0.088	-19.043	0.028	0.264	0.942	0.047	0.150	8.537	0.004	0.679	-0.051	0.024	0.300	
118	-2.931	0.056	0.244	-16.534	0.079	0.164	1.872	0.047	0.289	11.817	0.000	0.991	-0.002	0.004	0.757	
146	-3.121	0.056	0.112	-18.215	0.021	0.332	0.840	0.001	0.889	0.862	0.008	0.556	0.074	0.002	0.795	
153+132	-3.173	0.030	0.247	-12.808	0.024	0.306	0.850	0.043	0.166	8.079	0.010	0.518	-0.078	0.046	0.155	
105	-3.066	0.051	0.140	-17.240	0.083	0.058	1.589	0.076	0.070	10.910	0.035	0.221	-0.151	0.054	0.128	
141+179	-3.221	0.106	0.028	-23.699	0.063	0.093	1.370	0.061	0.098	9.531	0.003	0.736	-0.040	0.026	0.289	
137+176+130	-3.551	0.037	0.348	-18.753	0.028	0.413	1.564	0.016	0.538	9.697	0.005	0.726	0.088	0.005	0.732	
163+138	-3.231	0.055	0.115	-17.146	0.041	0.175	1.109	0.052	0.127	8.797	0.010	0.518	-0.077	0.044	0.160	

		Ozone			Carbon	Monoxide		NOx			Wind S	peed		Inv Ter	nperature	
CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\frac{WS}{R^2}$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
158	-3.221	0.074	0.068	-18.967	0.052	0.126	1.195	0.074	0.068	10.028	0.016	0.403	-0.095	0.044	0.163	4.80E+0
178+129	-3.371	0.050	0.141	-17.761	0.056	0.119	1.401	0.061	0.103	10.300	0.012	0.469	-0.095	0.012	0.473	2.86E+0
187+182	-3.520	0.113	0.023	-20.635	0.075	0.065	1.263	0.097	0.036	10.105	0.001	0.860	-0.018	0.001	0.881	-4.58E+
183	-3.461	0.110	0.025	-22.462	0.098	0.034	1.589	0.119	0.019	12.357	0.002	0.792	-0.029	0.005	0.635	1.61E+0
128	-3.469	0.005	0.639	-6.029	0.041	0.181	1.241	0.047	0.152	9.450	0.045	0.160	-0.188	0.007	0.582	2.28E+0
185	-3.501	0.000	0.932	-0.884	0.039	0.192	1.008	0.031	0.250	6.289	0.005	0.646	0.052	0.014	0.435	-2.70E+
174	-3.751	0.097	0.035	-19.841	0.077	0.063	1.322	0.081	0.055	9.617	0.001	0.843	-0.021	0.012	0.469	2.29E+0
177	-3.611	0.064	0.089	-17.973	0.060	0.102	1.298	0.071	0.073	10.020	0.003	0.698	-0.045	0.013	0.447	2.68E+0
202+171+156	-3.289	0.042	0.172	-13.602	0.041	0.178	1.004	0.053	0.124	8.069	0.008	0.562	-0.063	0.006	0.597	1.74E+0
180	-3.889	0.054	0.119	-16.533	0.063	0.094	1.329	0.075	0.066	10.268	0.001	0.814	-0.027	0.005	0.646	1.62E+0
199	-4.237	0.054	0.119	-14.917	0.066	0.086	1.226	0.055	0.116	7.944	0.003	0.707	-0.040	0.010	0.504	-2.13E+
170+190	-4.075	0.022	0.323	-10.858	0.044	0.161	1.147	0.054	0.121	8.927	0.007	0.590	-0.064	0.026	0.287	3.84E+0
201	-3.769	0.025	0.290	-11.749	0.046	0.151	1.189	0.043	0.166	8.088	0.000	0.961	-0.006	0.038	0.197	-4.70E+
203+196	-4.207	0.037	0.200	-13.558	0.059	0.103	1.282	0.059	0.105	9.001	0.000	0.950	-0.007	0.032	0.235	-4.14E+
195+208	-4.357	0.000	0.898	1.691	0.007	0.580	0.548	0.002	0.786	1.899	0.004	0.676	0.060	0.023	0.313	-4.36E+
194	-4.698	0.017	0.385	-10.280	0.038	0.193	1.148	0.047	0.148	8.987	0.002	0.783	-0.035	0.007	0.580	-2.16E+
206	-4.974	0.009	0.524	8.918	0.006	0.622	0.518	0.002	0.800	1.876	0.000	0.971	-0.005	0.044	0.163	-6.37E+
∑PCBs (incl. 8+5)		0.041	0.179	-12.514	0.053	0.124	1.068	0.045	0.156	6.977	0.003	0.710	-0.038	0.033	0.230	-6.58E-
∑PCBs (w/o 8+5)		0.043	0.165	-12.975	0.053	0.123	1.076	0.046	0.151	7.099	0.003	0.710	-0.038	0.033	0.231	-6.60E-

Table I-2. Summary of particle phase linear regressions (continued).

	[	Sine W	ind Direction	on	Cosine	Wind Dire	ction	Station	Pressure		Relative	e Humidity		Solar R	adiation	
CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
8+5	-0.808	0.000	0.966	0.013	0.003	0.760	-0.073	0.017	0.467	-0.018	0.006	0.662	-0.005	0.001	0.897	-0.220
18	-1.115	0.004	0.689	-0.110	0.001	0.860	0.041	0.000	0.952	-0.001	0.011	0.482	-0.008	0.016	0.405	-1.307

		Sine W	ind Direction	on	Cosine	Wind Dire	ction	Station	Pressure		Relativ	e Humidity	•	Solar R	Radiation	
CONGENER GROUP	log p	R <sup>2</sup>	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slo
17+15	-1.155	0.011	0.607	-0.221	0.001	0.883	0.047	0.006	0.706	-0.016	0.005	0.741	-0.005	0.008	0.675	-0.
16+32	-1.275	0.001	0.830	-0.061	0.004	0.671	0.099	0.000	0.941	0.002	0.014	0.432	-0.009	0.000	0.894	-0
31	-1.467	0.011	0.491	-0.149	0.002	0.760	0.056	0.000	0.965	0.001	0.011	0.490	-0.006	0.009	0.538	-0
28	-1.474	0.003	0.713	-0.081	0.000	0.973	-0.006	0.008	0.556	-0.012	0.004	0.676	-0.004	0.018	0.372	-1
21+33+53	-1.564	0.000	0.976	0.012	0.006	0.712	-0.107	0.009	0.649	-0.017	0.035	0.362	-0.012	0.000	0.933	0
22	-1.594	0.002	0.777	-0.072	0.001	0.889	0.030	0.000	0.939	-0.002	0.001	0.855	-0.002	0.030	0.251	-1
45	-1.655	0.000	0.979	-0.009	0.007	0.667	-0.115	0.009	0.617	-0.016	0.022	0.426	-0.010	0.000	0.933	0
46	-1.700	0.001	0.906	-0.050	0.009	0.640	-0.145	0.030	0.395	-0.034	0.000	0.984	0.000	0.014	0.570	-
52+43	-1.801	0.006	0.606	-0.119	0.030	0.247	0.223	0.006	0.619	0.010	0.013	0.444	-0.007	0.022	0.329	-
49	-1.781	0.005	0.626	-0.108	0.019	0.365	0.169	0.000	0.990	0.000	0.011	0.490	-0.006	0.015	0.415	-
47+48	-1.821	0.000	0.933	0.020	0.009	0.526	0.125	0.000	0.897	0.003	0.001	0.824	-0.002	0.008	0.561	-
44	-1.901	0.005	0.640	-0.106	0.025	0.293	0.199	0.000	0.980	-0.001	0.004	0.671	-0.004	0.008	0.554	-
37+42	-1.905	0.000	0.950	-0.017	0.018	0.379	0.203	0.005	0.652	-0.011	0.000	0.974	0.000	0.021	0.340	-
41+71	-1.971	0.003	0.734	-0.093	0.028	0.265	0.255	0.000	0.962	0.001	0.019	0.364	-0.010	0.002	0.800	-
64	-1.891	0.000	0.952	-0.014	0.011	0.479	0.140	0.000	0.889	-0.003	0.010	0.519	-0.006	0.004	0.678	-
40	-2.012	0.212	0.018	1.342	0.010	0.623	0.218	0.156	0.046	0.112	0.004	0.773	0.006	0.026	0.428	-
74	-2.145	0.003	0.706	-0.104	0.039	0.187	0.302	0.007	0.578	0.014	0.003	0.722	-0.004	0.004	0.688	-
70+76	-2.269	0.004	0.677	-0.108	0.084	0.051	0.418	0.022	0.325	0.023	0.017	0.396	-0.009	0.005	0.651	-
66+95	-2.214	0.004	0.674	-0.099	0.074	0.067	0.356	0.019	0.358	0.019	0.018	0.378	-0.008	0.013	0.458	-
91	-2.322	0.000	0.910	0.037	0.035	0.248	0.289	0.002	0.814	-0.007	0.001	0.818	-0.003	0.063	0.118	-
56+60+89	-2.375	0.004	0.660	-0.116	0.088	0.045	0.434	0.029	0.259	0.026	0.040	0.184	-0.014	0.000	0.963	-
92+84	-2.414	0.000	0.986	-0.004	0.089	0.044	0.402	0.014	0.427	0.017	0.026	0.289	-0.010	0.006	0.616	-
101	-2.478	0.007	0.594	-0.122	0.112	0.023	0.426	0.080	0.056	0.038	0.033	0.226	-0.011	0.016	0.400	-
99	-2.531	0.002	0.763	-0.077	0.070	0.075	0.376	0.013	0.453	0.017	0.010	0.507	-0.007	0.019	0.359	-
83	-2.574	0.012	0.502	-0.199	0.049	0.166	0.324	0.002	0.804	0.006	0.001	0.869	-0.002	0.050	0.159	-
97	-2.614	0.004	0.688	-0.094	0.132	0.013	0.472	0.105	0.028	0.044	0.049	0.139	-0.014	0.010	0.505	-
87+81	-2.649	0.005	0.746	0.147	0.059	0.231	0.397	0.007	0.688	0.018	0.013	0.585	-0.008	0.010	0.632	-
85+136	-2.634	0.025	0.296	-0.264	0.046	0.154	0.301	0.011	0.482	-0.016	0.000	0.970	0.000	0.001	0.808	-
110+77	-2.744	0.001	0.885	-0.037	0.118	0.020	0.486	0.081	0.055	0.043	0.051	0.131	-0.015	0.000	0.902	0

		Sine W	ind Directi	on	Cosine	Wind Dire	ction	Station	Pressure		Relativ	e Humidity	,	Solar F	Radiation	
CONGENER GROUP	log p	R <sup>2</sup>	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	1
82	-2.544	0.000	0.913	-0.031	0.038	0.194	0.306	0.001	0.832	-0.005	0.002	0.775	-0.003	0.006	0.609	
151	-2.758	0.004	0.677	-0.092	0.089	0.044	0.367	0.042	0.170	0.027	0.024	0.305	-0.009	0.024	0.302	
135+144+147+124	-2.948	0.000	0.931	-0.023	0.061	0.097	0.366	0.050	0.136	-0.006	0.007	0.568	-0.006	0.059	0.103	
149+123+107	-2.955	0.001	0.821	0.061	0.099	0.033	0.472	0.057	0.110	0.038	0.022	0.325	-0.011	0.012	0.478	
118	-2.931	0.002	0.830	0.093	0.073	0.181	0.420	0.016	0.539	0.026	0.006	0.715	-0.005	0.013	0.584	
146	-3.121	0.003	0.698	0.107	0.033	0.228	0.278	0.001	0.879	-0.004	0.000	0.963	-0.001	0.043	0.169	
153+132	-3.173	0.000	0.931	-0.023	0.089	0.044	0.440	0.037	0.198	0.030	0.022	0.331	-0.010	0.002	0.793	
105	-3.066	0.000	0.931	-0.025	0.062	0.103	0.369	0.006	0.628	0.012	0.000	0.901	0.001	0.013	0.467	
141+179	-3.221	0.000	0.958	0.014	0.058	0.108	0.350	0.021	0.340	0.022	0.004	0.697	-0.004	0.044	0.162	
137+176+130	-3.551	0.014	0.565	0.348	0.036	0.351	0.415	0.003	0.782	-0.016	0.007	0.684	-0.008	0.006	0.700	
163+138	-3.231	0.001	0.864	0.045	0.081	0.055	0.415	0.033	0.226	0.028	0.008	0.559	-0.006	0.014	0.443	
158	-3.221	0.000	0.968	-0.010	0.075	0.065	0.383	0.047	0.150	0.032	0.005	0.635	-0.005	0.021	0.343	
178+129	-3.371	0.000	0.898	0.038	0.065	0.092	0.403	0.013	0.465	0.019	0.009	0.538	-0.007	0.008	0.555	
187+182	-3.520	0.003	0.742	-0.073	0.076	0.063	0.340	0.008	0.559	0.011	0.013	0.450	-0.007	0.020	0.344	
183	-3.461	0.000	0.988	-0.004	0.067	0.082	0.352	0.015	0.415	0.018	0.010	0.520	-0.006	0.029	0.260	
128	-3.469	0.001	0.858	-0.054	0.031	0.246	0.289	0.004	0.676	0.011	0.013	0.454	-0.009	0.005	0.654	
185	-3.501	0.019	0.369	-0.222	0.034	0.228	0.250	0.002	0.801	-0.006	0.016	0.402	-0.008	0.002	0.764	
174	-3.751	0.001	0.826	0.051	0.059	0.104	0.310	0.011	0.489	0.014	0.008	0.548	-0.006	0.025	0.290	
177	-3.611	0.000	0.894	0.034	0.056	0.114	0.335	0.014	0.429	0.018	0.011	0.495	-0.007	0.013	0.457	
202+171+156	-3.289	0.002	0.762	-0.072	0.074	0.068	0.359	0.001	0.838	0.004	0.008	0.548	-0.006	0.000	0.992	
180	-3.889	0.000	0.971	-0.009	0.067	0.082	0.368	0.007	0.586	0.012	0.007	0.578	-0.006	0.008	0.565	
199	-4.237	0.001	0.820	0.052	0.054	0.121	0.297	0.000	0.964	-0.001	0.000	0.920	-0.001	0.001	0.840	
170+190	-4.075	0.000	0.927	-0.024	0.069	0.078	0.383	0.002	0.791	0.006	0.003	0.703	-0.004	0.001	0.841	
201	-3.769	0.003	0.739	-0.088	0.055	0.119	0.344	0.015	0.425	-0.019	0.004	0.684	-0.004	0.006	0.612	
203+196	-4.207	0.001	0.832	-0.054	0.055	0.116	0.331	0.012	0.466	-0.016	0.002	0.747	-0.003	0.001	0.812	
195+208	-4.357	0.000	0.900	0.039	0.086	0.048	0.512	0.022	0.326	-0.027	0.019	0.357	-0.012	0.066	0.085	
194	-4.698	0.001	0.870	0.046	0.031	0.243	0.275	0.010	0.514	-0.016	0.004	0.660	-0.005	0.010	0.503	
206	-4.974	0.002	0.774	-0.096	0.043	0.168	0.383	0.053	0.123	-0.045	0.006	0.603	-0.007	0.103	0.029	
$\sum$ PCBs (incl. 8+5)		0.064	0.091	0.313	0.051	0.132	0.202	0.033	0.225	0.001	0.001	0.813	0.001	0.000	0.905	

			Sine Wi			Cosine	Wind Direc	ction	Station	Pressure		Relative	e Humidity		Solar R	adiation	
	CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope	$\mathbb{R}^2$	p-value	Slope
-	∑PCBs (w/o 8+5)		0.067	0.083	0.323	0.054	0.120	0.209	0.033	0.226	0.001	0.001	0.811	0.001	0.000	0.897	0.182

Table I-3. Summary of multiple linear regressions including temperature (1/T) and Julian Date (JD).

CONGENER GROUP	log p	R2	p-value	a <sub>1</sub> (1/T)	p-value	a2 (JD)	p-value	Intercept	p-value
18	-1.115	0.360	0.0001	-5.60E+03	<.0001	-5.05E-04	0.321	26.1	<.0001
17+15	-1.155	0.410	<.0001	-5.40E+03	<.0001	-8.00E-04	0.103	26.0	<.0001
16+32	-1.275	0.402	<.0001	-6.18E+03	<.0001	-5.33E-04	0.298	28.2	<.0001
31	-1.467	0.518	<.0001	-6.75E+03	<.0001	-8.41E-04	0.069	31.9	<.0001
28	-1.474	0.474	<.0001	-5.40E+03	<.0001	-6.41E-04	0.109	11.9	0.1476
21+33+53	-1.564	0.498	<.0001	-7.47E+03	<.0001	-9.65E-04	0.191	34.5	<.0001
22	-1.594	0.406	<.0001	-6.95E+03	<.0001	-5.80E-04	0.309	30.5	<.0001
45	-1.655	0.497	<.0001	-6.10E+03	<.0001	-4.32E-04	0.295	25.5	<.0001
46	-1.700	0.565	0.0002	-9.96E+03	<.0001	-9.34E-04	0.633	40.3	0.0031
52+43	-1.801	0.611	<.0001	-6.29E+03	<.0001	-6.34E-04	0.070	29.8	<.0001
49	-1.781	0.576	<.0001	-6.11E+03	<.0001	-5.43E-04	0.131	27.6	<.0001
47+48	-1.821	0.525	<.0001	-5.93E+03	<.0001	-7.00E-04	0.078	27.0	<.0001
44	-1.901	0.608	<.0001	-6.62E+03	<.0001	-6.48E-04	0.079	30.4	<.0001
37+42	-1.905	0.560	<.0001	-7.13E+03	<.0001	-4.44E-04	0.293	30.2	<.0001
41+71	-1.971	0.537	<.0001	-7.33E+03	<.0001	-6.84E-04	0.144	31.9	<.0001
64	-1.891	0.565	<.0001	-6.57E+03	<.0001	-5.79E-04	0.142	28.5	<.0001
40	-2.012	0.553	<.0001	-8.49E+03	<.0001	7.71E-04	0.358	28.1	0.0007
74	-2.145	0.635	<.0001	-6.78E+03	<.0001	-6.22E-04	0.080	29.7	<.0001
70+76	-2.269	0.658	<.0001	-6.88E+03	<.0001	-5.75E-04	0.091	30.7	<.0001
66+95	-2.214	0.666	<.0001	-6.83E+03	<.0001	-5.89E-04	0.077	31.7	<.0001
91	-2.322	0.699	<.0001	-6.39E+03	<.0001	-5.75E-04	0.048	27.9	<.0001
56+60+89	-2.375	0.611	<.0001	-7.32E+03	<.0001	-4.51E-04	0.248	31.3	<.0001

CONGENER GROUP	log p	R2	p-value	a <sub>1</sub> (1/T)	p-value	a2 (JD)	p-value	Intercept	p-value
92+84	-2.414	0.552	<.0001	-7.07E+03	<.0001	-3.58E-04	0.397	30.5	<.0001
101	-2.478	0.668	<.0001	-6.91E+03	<.0001	-3.90E-04	0.230	30.5	<.0001
99	-2.531	0.655	<.0001	-7.35E+03	<.0001	-2.17E-04	0.534	30.3	<.0001
83	-2.574	0.683	<.0001	-7.45E+03	<.0001	-6.36E-05	0.851	28.2	<.0001
97	-2.614	0.564	<.0001	-6.66E+03	<.0001	-1.85E-04	0.629	27.2	<.0001
87+81	-2.649	0.617	<.0001	-8.01E+03	<.0001	-6.64E-04	0.288	34.9	<.0001
85+136	-2.634	0.622	<.0001	-8.48E+03	<.0001	1.17E-03	0.010	27.7	<.0001
110+77	-2.744	0.524	<.0001	-7.64E+03	<.0001	-1.35E-04	0.775	31.8	<.0001
82	-2.544	0.535	<.0001	-1.04E+04	<.0001	-5.87E-04	0.362	40.6	<.0001
151	-2.758	0.684	<.0001	-6.91E+03	<.0001	-2.83E-04	0.361	27.7	<.0001
135+144+147+124	-2.948	0.684	<.0001	-7.41E+03	<.0001	-4.70E-04	0.165	30.6	<.0001
149+123+107	-2.955	0.525	<.0001	-7.43E+03	<.0001	-2.16E-04	0.639	30.4	<.0001
118	-2.931	0.650	<.0001	-8.64E+03	<.0001	-1.05E-03	0.097	38.5	<.0001
146	-3.121	0.741	<.0001	-9.24E+03	<.0001	2.66E-07	0.999	34.3	<.0001
153+132	-3.173	0.715	<.0001	-8.83E+03	<.0001	-3.86E-04	0.295	36.0	<.0001
105	-3.066	0.623	<.0001	-9.72E+03	<.0001	-2.55E-04	0.609	37.7	<.0001
141	-3.221	0.679	<.0001	-8.65E+03	<.0001	-5.16E-04	0.196	34.4	<.0001
137+176+130	-3.551	0.553	<.0001	-1.11E+04	<.0001	-3.24E-03	0.003	53.4	<.0001
163+138	-3.231	0.731	<.0001	-9.97E+03	<.0001	-5.20E-04	0.198	40.7	<.0001
158	-3.221	0.731	<.0001	-9.85E+03	<.0001	-1.61E-04	0.678	36.7	<.0001
178+129	-3.371	0.701	<.0001	-9.45E+03	<.0001	-9.56E-04	0.030	38.4	<.0001
187+182	-3.520	0.830	<.0001	-9.80E+03	<.0001	-1.42E-04	0.618	36.8	<.0001
183	-3.461	0.788	<.0001	-1.04E+04	<.0001	-7.42E-05	0.832	37.7	<.0001
128	-3.469	0.587	<.0001	-1.16E+04	<.0001	-3.06E-04	0.633	42.3	<.0001
185	-3.501	0.816	<.0001	-1.03E+04	<.0001	9.90E-05	0.755	34.9	<.0001
174	-3.751	0.782	<.0001	-1.03E+04	<.0001	-2.75E-04	0.439	38.2	<.0001
177	-3.611	0.774	<.0001	-1.04E+04	<.0001	-3.77E-04	0.304	38.4	<.0001
202+171+156	-3.289	0.836	<.0001	-1.22E+04	<.0001	2.90E-05	0.934	44.2	<.0001
180	-3.889	0.817	<.0001	-1.30E+04	<.0001	7.77E-06	0.984	46.8	<.0001
199	-4.237	0.865	<.0001	-1.22E+04	<.0001	2.61E-04	0.404	41.3	<.0001

CONGENER GROUP	log p	R2	p-value	a <sub>1</sub> (1/T)	p-value	a2 (JD)	p-value	Intercept	p-value
170+190	-4.075	0.825	<.0001	-1.34E+04	<.0001	-1.69E-04	0.672	47.5	<.0001
201	-3.769	0.885	<.0001	-1.46E+04	<.0001	4.39E-04	0.198	51.1	<.0001
203+196	-4.207	0.897	<.0001	-1.46E+04	<.0001	4.80E-04	0.137	50.9	<.0001
195+208	-4.357	0.593	<.0001	-1.30E+04	<.0001	-3.29E-04	0.640	46.8	<.0001
194	-4.698	0.874	<.0001	-1.42E+04	<.0001	2.77E-05	0.937	48.7	<.0001
206	-4.974	0.762	<.0001	-1.35E+04	<.0001	2.21E-04	0.644	46.1	<.0001

Table I-4a. Summary of multiple linear regressions including temperature (1/T) and ozone  $(O_3)$ .

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (O <sub>3</sub> )	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
18	-1.1147	0.4223	<.0001	-23.931	0.0232	-7.3E+03	<.0001	30.7	<.0001
17+15	-1.1547	0.4376	<.0001	-20.503	0.0337	-6.9E+03	<.0001	28.2	<.0001
16+32	-1.2747	0.4241	<.0001	-17.508	0.1043	-7.5E+03	<.0001	31.0	<.0001
31	-1.4674	0.5156	<.0001	-17.309	0.0785	-8.1E+03	<.0001	33.6	<.0001
28	-1.4745	0.4723	<.0001	-13.254	0.1196	-6.5E+03	<.0001	27.3	<.0001
21+33+53	-1.5645	0.4862	<.0001	-10.217	0.3051	-7.6E+03	<.0001	31.1	<.0001
22	-1.5945	0.3999	<.0001	-9.7342	0.4224	-7.8E+03	<.0001	31.2	<.0001
45	-1.6554	0.5107	<.0001	-13.041	0.1343	-7.1E+03	<.0001	27.5	<.0001
46	-1.7000	0.5715	0.0002	-9.6433	0.475	-1.1E+04	0.0001	38.5	<.0001
52+43	-1.8011	0.5943	<.0001	-9.4149	0.2097	-7.1E+03	<.0001	30.1	<.0001
49	-1.7813	0.5864	<.0001	-13.791	0.0695	-7.2E+03	<.0001	29.4	<.0001
47+48	-1.8213	0.5405	<.0001	-17.674	0.035	-7.3E+03	<.0001	29.3	<.0001
44	-1.9013	0.5892	<.0001	-8.7035	0.2727	-7.4E+03	<.0001	30.5	<.0001
37+42	-1.9049	0.5725	<.0001	-13.438	0.1318	-8.2E+03	<.0001	32.3	<.0001
41+71	-1.9713	0.5343	<.0001	-13.825	0.1652	-8.5E+03	<.0001	33.3	<.0001
64	-1.8913	0.5477	<.0001	-6.3008	0.4565	-7.2E+03	<.0001	28.3	<.0001
40	-2.0122	0.5440	<.0001	5.74789	0.6115	-8.5E+03	<.0001	31.5	<.0001
74	-2.1447	0.6183	<.0001	-8.656	0.2565	-7.5E+03	<.0001	29.9	<.0001
70+76	-2.2691	0.6381	<.0001	-5.7214	0.4353	-7.4E+03	<.0001	30.3	<.0001

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (O <sub>3</sub> )	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
66+95	-2.2140	0.6410	<.0001	-3.599	0.6173	-7.2E+03	<.0001	30.7	<.0001
91	-2.3217	0.6703	<.0001	-3.2995	0.6019	-6.8E+03	<.0001	26.9	<.0001
56+60+89	-2.3747	0.5985	<.0001	0.95863	0.9088	-7.4E+03	<.0001	29.6	<.0001
92+84	-2.4145	0.5438	<.0001	-0.7736	0.9315	-7.2E+03	<.0001	29.6	<.0001
101	-2.4777	0.6575	<.0001	-2.5566	0.7134	-7.2E+03	<.0001	29.9	<.0001
99	-2.5314	0.6568	<.0001	-5.6643	0.4441	-7.8E+03	<.0001	31.1	<.0001
83	-2.5745	0.6828	<.0001	-0.6874	0.9262	-7.5E+03	<.0001	28.2	<.0001
97	-2.6145	0.5617	<.0001	1.42647	0.8609	-6.6E+03	<.0001	26.2	<.0001
87+81	-2.6487	0.6022	<.0001	-1.1045	0.8962	-7.7E+03	<.0001	30.9	<.0001
85+136	-2.6345	0.5628	<.0001	-8.9681	0.3688	-8.8E+03	<.0001	33.8	<.0001
110+77	-2.7445	0.5264	<.0001	5.07814	0.6126	-7.3E+03	<.0001	30.1	<.0001
82	-2.5445	0.5279	<.0001	6.3758	0.6422	-1.0E+04	<.0001	37.0	<.0001
151	-2.7581	0.6798	<.0001	-3.5807	0.5874	-7.2E+03	<.0001	27.7	<.0001
135+144+147+124	-2.9481	0.6693	<.0001	-1.7073	0.8145	-7.7E+03	<.0001	29.5	<.0001
149+123+107	-2.9549	0.5227	<.0001	2.25772	0.8181	-7.3E+03	<.0001	29.1	<.0001
118	-2.9310	0.6157	<.0001	-1.5085	0.8617	-8.1E+03	<.0001	32.1	<.0001
146	-3.1211	0.7408	<.0001	-0.4323	0.954	-9.3E+03	<.0001	34.4	<.0001
153+132	-3.1735	0.7071	<.0001	-0.2605	0.9737	-9.0E+03	<.0001	34.8	<.0001
105	-3.0664	0.6208	<.0001	0.07927	0.9941	-9.8E+03	<.0001	36.9	<.0001
141	-3.2211	0.6672	<.0001	-3.9292	0.6455	-9.1E+03	<.0001	33.8	<.0001
137+176+130	-3.5511	0.3914	0.0006	-0.5093	0.9739	-9.2E+03	0.0005	32.4	0.0006
163+138	-3.2311	0.7204	<.0001	-2.7765	0.7483	-1.0E+04	<.0001	39.7	<.0001
158	-3.2211	0.7319	<.0001	-4.1531	0.6139	-1.0E+04	<.0001	37.2	<.0001
178+129	-3.3710	0.6689	<.0001	-7.7921	0.4206	-1.0E+04	<.0001	37.4	<.0001
187+182	-3.5199	0.8482	<.0001	-12.686	0.0292	-1.1E+04	<.0001	39.9	<.0001
183	-3.4610	0.7956	<.0001	-8.8988	0.2271	-1.1E+04	<.0001	39.8	<.0001
128	-3.4692	0.5866	<.0001	6.56382	0.6315	-1.1E+04	<.0001	39.8	<.0001
185	-3.5010	0.8248	<.0001	-9.6779	0.1451	-1.1E+04	<.0001	37.7	<.0001
174	-3.7510	0.7825	<.0001	-6.1681	0.4132	-1.1E+04	<.0001	38.9	<.0001
177	-3.6110	0.7718	<.0001	-5.9971	0.437	-1.1E+04	<.0001	39.0	<.0001
202+171+156	-3.2885	0.8430	<.0001	-9.9849	0.1727	-1.3E+04	<.0001	46.8	<.0001

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (O <sub>3</sub> )	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
180	-3.8895	0.8253	<.0001	-11.768	0.1597	-1.4E+04	<.0001	49.8	<.0001
199	-4.2373	0.8822	<.0001	-16.031	0.0124	-1.3E+04	<.0001	46.3	<.0001
170+190	-4.0750	0.8246	<.0001	-2.0348	0.8111	-1.4E+04	<.0001	47.5	<.0001
201	-3.7685	0.8930	<.0001	-15.052	0.0346	-1.6E+04	<.0001	56.4	<.0001
203+196	-4.2073	0.9027	<.0001	-14.553	0.0312	-1.5E+04	<.0001	56.2	<.0001
195+208	-4.3573	0.5995	<.0001	13.7645	0.3544	-1.2E+04	<.0001	42.2	<.0001
194	-4.6975	0.8780	<.0001	-8.5341	0.2461	-1.5E+04	<.0001	51.0	<.0001
206	-4.9736	0.7609	<.0001	-3.0499	0.7614	-1.4E+04	<.0001	47.6	<.0001

Table I-4b. Summary	1	ı	1	1	1	1	· ·	1	Ι.,
CONGENER GROUP	log p	R <sup>2</sup>	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
18	-1.1147	0.4751	<.0001	-0.2559	0.0026	-5.1E+03	<.0001	23.2	<.0001
17+15	-1.1547	0.5247	<.0001	-0.2648	0.0009	-4.7E+03	<.0001	21.4	<.0001
16+32	-1.2747	0.5372	<.0001	-0.2865	0.0007	-5.6E+03	<.0001	25.0	<.0001
31	-1.4674	0.6074	<.0001	-0.2633	0.0007	-6.3E+03	<.0001	27.8	<.0001
28	-1.4745	0.5969	<.0001	-0.2408	0.0003	-5.0E+03	<.0001	22.6	<.0001
21+33+53	-1.5645	0.6295	<.0001	-0.2695	0.0011	-6.3E+03	<.0001	27.3	<.0001
22	-1.5945	0.5251	<.0001	-0.3013	0.0015	-6.3E+03	<.0001	27.1	<.0001
45	-1.6554	0.6225	<.0001	-0.2414	0.0004	-5.6E+03	<.0001	22.8	<.0001
46	-1.7000	0.6553	<.0001	-0.2626	0.0292	-8.5E+03	0.0001	32.1	<.0001
52+43	-1.8011	0.6731	<.0001	-0.1891	0.0013	-6.0E+03	<.0001	26.7	<.0001
49	-1.7813	0.6560	<.0001	-0.197	0.001	-5.7E+03	<.0001	24.8	<.0001
47+48	-1.8213	0.5975	<.0001	-0.2102	0.0017	-5.6E+03	<.0001	23.6	<.0001
44	-1.9013	0.6819	<.0001	-0.2095	0.0007	-6.3E+03	<.0001	27.2	<.0001
37+42	-1.9049	0.6235	<.0001	-0.1947	0.0064	-6.7E+03	<.0001	27.8	<.0001
41+71	-1.9713	0.6293	<.0001	-0.2604	0.0008	-6.8E+03	<.0001	28.4	<.0001
64	-1.8913	0.6501	<.0001	-0.2174	0.0009	-6.2E+03	<.0001	25.5	<.0001
40	-2.0122	0.6302	<.0001	-0.2442	0.0111	-8.4E+03	<.0001	32.3	<.0001

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
74	-2.1447	0.6852	<.0001	-0.1814	0.0026	-6.5E+03	<.0001	26.7	<.0001
70+76	-2.2691	0.7077	<.0001	-0.175	0.0023	-6.6E+03	<.0001	27.9	<.0001
66+95	-2.2140	0.7171	<.0001	-0.1767	0.0017	-6.5E+03	<.0001	28.9	<.0001
91	-2.3217	0.7430	<.0001	-0.1584	0.0013	-6.1E+03	<.0001	25.2	<.0001
56+60+89	-2.3747	0.6716	<.0001	-0.1879	0.0043	-7.0E+03	<.0001	28.9	<.0001
92+84	-2.4145	0.6488	<.0001	-0.2274	0.0011	-6.6E+03	<.0001	28.2	<.0001
101	-2.4777	0.7183	<.0001	-0.1556	0.0045	-6.6E+03	<.0001	28.5	<.0001
99	-2.5314	0.7078	<.0001	-0.1568	0.0077	-7.0E+03	<.0001	28.8	<.0001
83	-2.5745	0.7051	<.0001	-0.1016	0.089	-7.2E+03	<.0001	27.4	<.0001
97	-2.6145	0.6366	<.0001	-0.1774	0.0058	-6.2E+03	<.0001	25.7	<.0001
87+81	-2.6487	0.7061	<.0001	-0.2105	0.0028	-7.1E+03	<.0001	29.8	<.0001
85+136	-2.6345	0.5822	<.000	-0.1326	0.1041	-7.8E+03	<.0001	30.8	<.0001
110+77	-2.7445	0.6110	<.0001	-0.2265	0.0041	-7.1E+03	<.0001	30.2	<.0001
82	-2.5445	0.5798	<.0001	-0.2446	0.0264	-9.9E+03	<.0001	37.4	<.0001
151	-2.7581	0.7323	<.0001	-0.1435	0.006	-6.6E+03	<.0001	26.0	<.0001
135+144+147+124	-2.9481	0.7278	<.0001	-0.1616	0.0048	-7.1E+03	<.0001	28.2	<.0001
149+123+107	-2.9549	0.6160	<.0001	-0.2291	0.0029	-6.9E+03	<.0001	28.5	<.0001
118	-2.9310	0.7181	<.0001	-0.2177	0.0024	-7.5E+03	<.0001	30.9	<.0001
146	-3.1211	0.7899	<.0001	-0.1717	0.0035	-8.8E+03	<.0001	33.4	<.0001
153+132	-3.1735	0.7673	<.0001	-0.1881	0.0023	-8.4E+03	<.0001	33.7	<.0001
105	-3.0664	0.6813	<.0001	-0.2177	0.0088	-9.3E+03	<.0001	35.8	<.0001
141	-3.2211	0.7029	<.0001	-0.1505	0.0284	-8.4E+03	<.0001	31.9	<.0001
137+176+130	-3.5511	0.4652	<.0001	-0.2638	0.0507	-8.6E+03	0.0001	31.4	<.0001
163+138	-3.2311	0.7711	<.0001	-0.195	0.0042	-9.6E+03	<.0001	38.0	<.0001
158	-3.2211	0.7744	<.0001	-0.1756	0.0071	-9.4E+03	<.0001	35.2	<.0001
178+129	-3.3710	0.7113	<.0001	-0.187	0.0138	-9.3E+03	<.0001	34.6	<.0001
187+182	-3.5199	0.8697	<.0001	-0.1517	0.001	-9.5E+03	<.0001	35.9	<.0001
183	-3.4610	0.8236	<.0001	-0.1599	0.0065	-1.0E+04	<.0001	36.6	<.0001
128	-3.4692	0.6420	<.0001	-0.2608	0.015	-1.1E+04	<.0001	40.1	<.0001
185	-3.5010	0.8429	<.0001	-0.1369	0.0105	-1.0E+04	<.0001	34.5	<.0001
174	-3.7510	0.8105	<.0001	-0.1505	0.0124	-1.0E+04	<.0001	36.5	<.0001

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
177	-3.6110	0.8019	<.0001	-0.1527	0.0129	-1.0E+04	<.0001	36.8	<.0001
202+171+156	-3.2885	0.8732	<.0001	-0.1864	0.0012	-1.2E+04	<.0001	43.3	<.0001
180	-3.8895	0.8425	<.0001	-0.1676	0.0129	-1.3E+04	<.0001	45.9	<.0001
199	-4.2373	0.8974	<.0001	-0.1763	0.0006	-1.2E+04	<.0001	41.2	<.0001
170+190	-4.0750	0.8436	<.0001	-0.1483	0.0302	-1.3E+04	<.0001	46.2	<.0001
201	-3.7685	0.9067	<.0001	-0.1802	0.0016	-1.4E+04	<.0001	51.6	<.0001
203+196	-4.2073	0.9145	<.0001	-0.1701	0.0017	-1.4E+04	<.0001	51.6	<.0001
195+208	-4.3573	0.6029	<.0001	-0.1345	0.2726	-1.3E+04	<.0001	45.0	<.0001
194	-4.6975	0.8935	<.0001	-0.1542	0.0087	-1.4E+04	<.0001	48.0	<.0001
206	-4.9736	0.7771	<.0001	-0.1379	0.0899	-1.3E+04	<.0001	46.2	<.0001

Table I-4c. Summary of multiple linear regressions including temperature (1/T) and wind direction (WD).

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> sin(WD)	p-value	a <sub>2</sub> cos(WD)	p-value	a <sub>3</sub> (1/T)	p-value	intercept	p-value
18	-1.1147	0.3569	0.0005	0.1145	0.561	-0.1143	0.481	-5.77E+03	<.0001	24.6	<.0001
17+15	-1.1547	0.3839	0.0003	0.1360	0.457	-0.1051	0.487	-5.52E+03	<.0001	23.1	<.0001
16+32	-1.2747	0.3988	0.0001	0.1448	0.465	-0.1023	0.530	-6.41E+03	<.0001	26.8	<.0001
31	-1.4674	0.4847	<.0001	0.1080	0.553	-0.0750	0.616	-7.05E+03	<.0001	29.3	<.0001
28	-1.4745	0.4556	<.0001	0.0915	0.557	-0.1188	0.355	-5.58E+03	<.0001	23.8	<.0001
21+33+53	-1.5645	0.4832	0.0002	0.1035	0.600	-0.1211	0.433	-6.87E+03	<.0001	28.3	<.0001
22	-1.5945	0.4047	0.0001	0.1873	0.396	-0.0945	0.601	-7.25E+03	<.0001	29.1	<.0001
45	-1.6554	0.4999	<.0001	0.1388	0.383	-0.1051	0.422	-6.29E+03	<.0001	24.3	<.0001
46	-1.7000	0.5738	0.0009	0.1667	0.578	-0.1017	0.643	-9.90E+03	<.0001	35.9	<.0001
52+43	-1.8011	0.5805	<.0001	0.0540	0.695	-0.0296	0.794	-6.51E+03	<.0001	27.8	<.0001
49	-1.7813	0.5550	<.0001	0.0478	0.735	-0.0553	0.634	-6.27E+03	<.0001	25.8	<.0001
47+48	-1.8213	0.4968	<.0001	0.0494	0.751	-0.1056	0.412	-6.08E+03	<.0001	24.6	<.0001
44	-1.9013	0.5819	<.0001	0.0911	0.531	-0.0385	0.747	-6.88E+03	<.0001	28.5	<.0001
37+42	-1.9049	0.5728	<.0001	0.2379	0.143	-0.0549	0.678	-7.50E+03	<.0001	29.6	<.0001
41+71	-1.9713	0.5181	<.0001	0.1181	0.519	-0.0542	0.719	-7.62E+03	<.0001	30.0	<.0001

CONGENER GROUP	log p	R <sup>2</sup>	p-value	a <sub>1</sub> sin(WD)	p-value	a <sub>2</sub> cos(WD)	p-value	a <sub>3</sub> (1/T)	p-value	intercept	p-value
64	-1.8913	0.5511	<.0001	0.1205	0.433	-0.0655	0.604	-6.82E+03	<.0001	27.0	<.0001
40	-2.0122	0.5458	<.0001	0.0845	0.706	-0.0855	0.626	-8.98E+03	<.0001	33.2	<.0001
74	-2.1447	0.6093	<.0001	0.0609	0.663	-0.0475	0.680	-6.98E+03	<.0001	27.8	<.0001
70+76	-2.2691	0.6355	<.0001	0.0675	0.614	-0.0301	0.785	-7.10E+03	<.0001	29.0	<.0001
66+95	-2.2140	0.6417	<.0001	0.0695	0.596	-0.0253	0.815	-7.06E+03	<.0001	30.0	<.0001
91	-2.3217	0.6781	<.0001	0.0748	0.512	-0.0871	0.355	-6.56E+03	<.0001	26.1	<.0001
56+60+89	-2.3747	0.6056	<.0001	0.1276	0.401	-0.0193	0.877	-7.59E+03	<.0001	30.3	<.0001
92+84	-2.4145	0.5490	<.0001	0.1105	0.500	-0.0138	0.918	-7.29E+03	<.0001	29.8	<.0001
101	-2.4777	0.6575	<.0001	0.0270	0.831	-0.0313	0.764	-7.02E+03	<.0001	29.3	<.0001
99	-2.5314	0.6571	<.0001	-0.0095	0.944	-0.0857	0.440	-7.32E+03	<.0001	29.3	<.0001
83	-2.5745	0.6854	<.0001	0.0169	0.895	-0.0601	0.575	-7.43E+03	<.0001	27.9	<.0001
97	-2.6145	0.5620	<.0001	-0.0343	0.817	0.0081	0.947	-6.68E+03	<.0001	26.5	<.0001
87+81	-2.6487	0.6111	<.0001	0.0617	0.711	-0.0962	0.461	-7.57E+03	<.0001	30.5	<.0001
85+136	-2.6345	0.5616	<.0001	0.0952	0.600	-0.0993	0.507	-8.17E+03	<.0001	31.6	<.0001
110+77	-2.7445	0.5245	<.0001	-0.0364	0.842	0.0367	0.807	-7.67E+03	<.0001	31.4	<.0001
82	-2.5445	0.5451	<.0001	0.3184	0.199	-0.0476	0.814	-1.09E+04	<.0001	39.9	<.0001
151	-2.7581	0.6805	<.0001	0.0045	0.972	-0.0428	0.669	-7.00E+03	<.0001	26.8	<.0001
135+144+147+124	-2.9481	0.6719	<.0001	0.0122	0.927	-0.0656	0.547	-7.50E+03	<.0001	28.9	<.0001
149+123+107	-2.9549	0.5323	<.0001	-0.1496	0.400	-0.0501	0.731	-7.25E+03	<.0001	28.9	<.0001
118	-2.9310	0.6262	<.0001	0.0478	0.779	-0.1162	0.384	-7.93E+03	<.0001	31.4	<.0001
146	-3.1211	0.7431	<.0001	-0.0420	0.758	-0.0558	0.619	-9.13E+03	<.0001	33.9	<.0001
153+132	-3.1735	0.7078	<.0001	-0.0046	0.975	-0.0348	0.768	-8.90E+03	<.0001	34.6	<.0001
105	-3.0664	0.6250	<.0001	0.1234	0.521	0.0113	0.942	-9.96E+03	<.0001	37.4	<.0001
141	-3.2211	0.6749	<.0001	0.0354	0.818	-0.1336	0.294	-8.71E+03	<.0001	32.4	<.0001
137+176+130	-3.5511	0.3953	0.002	-0.0425	0.891	0.0997	0.680	-9.17E+03	0.0002	32.5	0.0002
163+138	-3.2311	0.7231	<.0001	-0.0335	0.831	-0.0833	0.520	-1.00E+04	<.0001	38.6	<.0001
158	-3.2211	0.7402	<.0001	-0.0814	0.582	-0.1294	0.290	-9.67E+03	<.0001	35.4	<.0001
178+129	-3.3710	0.6711	<.0001	-0.1624	0.352	-0.0167	0.906	-9.60E+03	<.0001	34.9	<.0001
187+182	-3.5199	0.8318	<.0001	0.0804	0.457	-0.0367	0.680	-9.93E+03	<.0001	36.7	<.0001
183	-3.4610	0.7901	<.0001	-0.0291	0.829	-0.0622	0.576	-1.04E+04	<.0001	37.1	<.0001
128	-3.4692	0.5902	<.0001	0.1845	0.454	-0.0184	0.927	-1.18E+04	<.0001	42.0	<.0001

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> sin(WD)	p-value	a <sub>2</sub> cos(WD)	p-value	a <sub>3</sub> (1/T)	p-value	intercept	p-value
185	-3.5010	0.8162	<.0001	0.0128	0.917	-0.0428	0.671	-1.03E+04	<.0001	35.1	<.0001
174	-3.7510	0.7805	<.0001	-0.0235	0.865	-0.0561	0.621	-1.03E+04	<.0001	37.0	<.0001
177	-3.6110	0.7696	<.0001	-0.0242	0.863	-0.0502	0.664	-1.05E+04	<.0001	37.2	<.0001
202+171+156	-3.2885	0.8369	<.0001	0.0359	0.790	0.0544	0.624	-1.23E+04	<.0001	44.6	<.0001
180	-3.8895	0.8205	<.0001	-0.0900	0.557	-0.0882	0.484	-1.28E+04	<.0001	46.1	<.0001
199	-4.2373	0.8650	<.0001	0.0906	0.453	-0.0422	0.670	-1.22E+04	<.0001	42.4	<.0001
170+190	-4.0750	0.8280	<.0001	-0.0745	0.628	-0.0932	0.463	-1.33E+04	<.0001	46.3	<.0001
201	-3.7685	0.8815	<.0001	0.0612	0.646	-0.0399	0.716	-1.45E+04	<.0001	52.7	<.0001
203+196	-4.2073	0.8917	<.0001	0.0469	0.710	-0.0430	0.679	-1.45E+04	<.0001	52.5	<.0001
195+208	-4.3573	0.6178	<.0001	0.4200	0.116	0.0865	0.690	-1.37E+04	<.0001	47.8	<.0001
194	-4.6975	0.8788	<.0001	0.1042	0.434	-0.1141	0.299	-1.43E+04	<.0001	48.9	<.0001
206	-4.9736	0.7699	<.0001	0.2202	0.222	-0.0521	0.724	-1.36E+04	<.0001	47.4	<.0001

Table I-4d. Summary of multiple linear regressions including temperature (1/T) and station pressure (P).

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (P)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
18	-1.1147	0.3452	0.0002	0.00518	0.7754	-5.9E+03	<.0001	19.7	0.2592
17+15	-1.1547	0.3685	0.0001	0.00365	0.8265	-5.6E+03	<.0001	19.6	0.2247
16+32	-1.2747	0.386	<.0001	0.00368	0.8404	-6.4E+03	<.0001	23.1	0.1912
31	-1.4674	0.4835	<.0001	0.01176	0.4809	-7.3E+03	<.0001	18.3	0.2551
28	-1.4745	0.4427	<.0001	0.00662	0.6458	-5.8E+03	<.0001	17.7	0.2025
21+33+53	-1.5645	0.4851	<.0001	0.01621	0.3199	-7.3E+03	<.0001	13.3	0.3958
22	-1.5945	0.3904	<.0001	0.00112	0.9562	-7.1E+03	<.0001	27.6	0.1605
45	-1.6554	0.4833	<.0001	0.00285	0.8464	-6.3E+03	<.0001	21.4	0.1332
46	-1.7000	0.5606	0.0003	-0.004	0.894	-9.8E+03	<.0001	39.5	0.2124
52+43	-1.8011	0.5878	<.0001	0.01215	0.3342	-6.8E+03	<.0001	16.5	0.1733
49	-1.7813	0.5572	<.0001	0.00942	0.4653	-6.5E+03	<.0001	17.1	0.1694
47+48	-1.8213	0.4908	<.0001	0.00766	0.5937	-6.3E+03	<.0001	17.6	0.2025
44	-1.9013	0.5827	<.0001	0.01003	0.4509	-7.1E+03	<.0001	19.0	0.1393
37+42	-1.9049	0.5581	<.0001	0.01449	0.3342	-7.6E+03	<.0001	15.4	0.284

CONGENER GROUP	log p	R <sup>2</sup>	p-value	a <sub>1</sub> (P)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
41+71	-1.9713	0.5158	<.0001	0.00988	0.5564	-7.8E+03	<.0001	20.6	0.2037
64	-1.8913	0.5435	<.0001	0.00591	0.6765	-6.9E+03	<.0001	21.2	0.1226
40	-2.0122	0.5444	<.0001	0.0098	0.5964	-9.2E+03	<.0001	24.1	0.1826
74	-2.1447	0.6122	<.0001	0.01031	0.4202	-7.2E+03	<.0001	18.2	0.1413
70+76	-2.2691	0.6379	<.0001	0.00938	0.4438	-7.3E+03	<.0001	20.2	0.0893
66+95	-2.2140	0.6452	<.0001	0.01024	0.3929	-7.3E+03	<.0001	20.3	0.0808
91	-2.3217	0.6715	<.0001	0.00684	0.5168	-6.7E+03	<.0001	19.7	0.0557
56+60+89	-2.3747	0.603	<.0001	0.00956	0.4922	-7.7E+03	<.0001	21.0	0.119
92+84	-2.4145	0.5447	<.0001	0.0045	0.7645	-7.3E+03	<.0001	25.2	0.085
101	-2.4777	0.6639	<.0001	0.01095	0.3438	-7.3E+03	<.0001	19.1	0.0872
99	-2.5314	0.6585	<.0001	0.01095	0.375	-7.7E+03	<.0001	19.5	0.1023
83	-2.5745	0.6833	<.0001	0.00314	0.7901	-7.5E+03	<.0001	25.1	0.0311
97	-2.6145	0.5667	<.0001	0.00958	0.4793	-7.0E+03	<.0001	17.7	0.1742
87+81	-2.6487	0.6121	<.0001	0.01197	0.3839	-7.9E+03	<.0001	19.6	0.1453
85+136	-2.6345	0.5566	<.0001	0.00808	0.6289	-8.4E+03	<.0001	24.0	0.1375
110+77	-2.7445	0.5267	<.0001	0.0089	0.595	-7.9E+03	<.0001	23.2	0.1523
82	-2.5445	0.5254	<.0001	-1E-05	0.9996	-1.1E+04	<.0001	38.7	0.0835
151	-2.7581	0.681	<.0001	0.00743	0.4995	-7.2E+03	<.0001	19.9	0.0634
135+144+147+124	-2.9481	0.6718	<.0001	0.00734	0.545	-7.7E+03	<.0001	22.3	0.0599
149+123+107	-2.9549	0.5267	<.0001	0.01032	0.5279	-7.8E+03	<.0001	20.1	0.2018
118	-2.9310	0.6213	<.0001	0.00962	0.4952	-8.3E+03	<.0001	22.8	0.1007
146	-3.1211	0.742	<.0001	0.00556	0.6563	-9.4E+03	<.0001	29.1	0.0184
153+132	-3.1735	0.7086	<.0001	0.00597	0.6497	-9.1E+03	<.0001	29.2	0.0242
105	-3.0664	0.6215	<.0001	-0.0046	0.7951	-9.7E+03	<.0001	41.1	0.0188
141	-3.2211	0.6657	<.0001	0.00248	0.8621	-8.9E+03	<.0001	30.4	0.0303
137+176+130	-3.5511	0.3918	0.0006	0.00394	0.8771	-9.2E+03	0.0002	28.7	0.2488
163+138	-3.2311	0.7208	<.0001	0.00582	0.6867	-1.0E+04	<.0001	33.6	0.0187
158	-3.2211	0.7305	<.0001	-0.0026	0.8515	-9.8E+03	<.0001	38.5	0.0053
178+129	-3.3710	0.6634	<.0001	-0.0007	0.9671	-9.8E+03	<.0001	36.1	0.023
187+182	-3.5199	0.8329	<.0001	0.00996	0.3266	-1.0E+04	<.0001	27.1	0.0085
183	-3.4610	0.7891	<.0001	0.00534	0.6663	-1.1E+04	<.0001	32.5	0.0085

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (P)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
128	-3.4692	0.5847	<.0001	-0.0049	0.8272	-1.2E+04	<.0001	45.9	0.0392
185	-3.5010	0.8209	<.0001	0.01239	0.2664	-1.1E+04	<.0001	23.8	0.0292
174	-3.7510	0.7818	<.0001	0.00928	0.4613	-10603	<.0001	28.7	0.0208
177	-3.6110	0.7731	<.0001	0.012	0.3612	-1.1E+04	<.0001	26.1	0.0457
202+171+156	-3.2885	0.8384	<.0001	0.0103	0.4028	-1.2E+04	<.0001	34.7	0.0048
180	-3.8895	0.8172	<.0001	0.00568	0.6878	-1.3E+04	<.0001	41.6	0.0035
199	-4.2373	0.8639	<.0001	0.00691	0.5336	-1.2E+04	<.0001	35.8	0.0015
170+190	-4.0750	0.826	<.0001	0.00859	0.5445	-1.4E+04	<.0001	39.0	0.006
201	-3.7685	0.8813	<.0001	0.00601	0.6221	-1.5E+04	<.0001	47.0	0.0002
203+196	-4.2073	0.8916	<.0001	0.00583	0.6145	-1.5E+04	<.0001	47.1	0.0001
195+208	-4.3573	0.591	<.0001	0.00243	0.9223	-1.3E+04	<.0001	43.5	0.0739
194	-4.6975	0.8829	<.0001	0.02108	0.0828	-1.5E+04	<.0001	29.3	0.0135
206	-4.9736	0.7661	<.0001	0.01671	0.3266	-1.4E+04	<.0001	31.0	0.0659

Table I-4e. Summary of multiple linear regressions including temperature (1/T) and relative humidity (RH).

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (RH)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
18	-1.1147	0.4818	<.0001	0.02212	0.002	-5.5E+03	<.0001	22.0	<.0001
17+15	-1.1547	0.4894	<.0001	0.01949	0.0041	-5.1E+03	<.0001	20.4	<.0001
16+32	-1.2747	0.5039	<.0001	0.02134	0.0032	-6.0E+03	<.0001	24.1	<.0001
31	-1.4674	0.615	<.0001	0.02283	0.0004	-6.7E+03	<.0001	26.6	<.0001
28	-1.4745	0.6018	<.0001	0.02062	0.0002	-5.3E+03	<.0001	21.5	<.0001
21+33+53	-1.5645	0.607	<.0001	0.01959	0.0028	-6.4E+03	<.0001	4.1	<.0001
22	-1.5945	0.4817	<.0001	0.02092	0.0103	-6.8E+03	<.0001	26.3	<.0001
45	-1.6554	0.6225	<.0001	0.02036	0.0004	-6.0E+03	<.0001	21.8	<.0001
46	-1.7000	0.6718	<.0001	0.02149	0.0168	-8.8E+03	<.0001	30.6	<.0001
52+43	-1.8011	0.6905	<.0001	0.01734	0.0004	-6.2E+03	<.0001	25.8	<.0001
49	-1.7813	0.7019	<.0001	0.01992	<.0001	-6.0E+03	<.0001	23.6	<.0001
47+48	-1.8213	0.6516	<.0001	0.02163	<.0001	-5.8E+03	<.0001	22.3	<.0001
44	-1.9013	0.6894	<.0001	0.01829	0.0004	-6.6E+03	<.0001	26.2	<.0001

CONGENER									
GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (RH)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
37+42	-1.9049	0.6422	<.0001	0.01832	0.0021	-7.0E+03	<.0001	26.7	<.0001
41+71	-1.9713	0.6369	<.0001	0.02265	0.0005	-7.2E+03	<.0001	27.2	<.0001
64	-1.8913	0.6548	<.0001	0.01872	0.0007	-6.5E+03	<.0001	24.5	<.0001
40	-2.0122	0.5974	<.0001	0.01526	0.0473	-8.6E+03	<.0001	30.9	<.0001
74	-2.1447	0.7235	<.0001	0.01862	0.0002	-6.7E+03	<.0001	25.6	<.0001
70+76	-2.2691	0.7376	<.0001	0.01745	0.0002	-6.8E+03	<.0001	26.9	<.0001
66+95	-2.2140	0.7325	<.0001	0.0163	0.0005	-6.8E+03	<.0001	27.9	<.0001
91	-2.3217	0.7506	<.0001	0.01401	0.0007	-6.4E+03	<.0001	24.4	<.0001
56+60+89	-2.3747	0.6591	<.0001	0.01442	0.01	-7.3E+03	<.0001	28.2	<.0001
92+84	-2.4145	0.6136	<.0001	0.01564	0.0094	-7.0E+03	<.0001	27.6	<.0001
101	-2.4777	0.753	<.0001	0.01639	0.0003	-6.8E+03	<.0001	27.4	<.0001
99	-2.5314	0.7557	<.0001	0.018	0.0002	-7.2E+03	<.0001	27.6	<.0001
83	-2.5745	0.7814	<.0001	0.01735	0.0001	-7.2E+03	<.0001	26.0	<.0001
97	-2.6145	0.6827	<.0001	0.01899	0.0003	-6.5E+03	<.0001	24.5	<.0001
87+81	-2.6487	0.6933	<.0001	0.01544	0.0056	-7.3E+03	<.0001	28.4	<.0001
85+136	-2.6345	0.6467	<.0001	0.02028	0.0021	-7.9E+03	<.0001	29.2	<.0001
110+77	-2.7445	0.6294	<.0001	0.02101	0.0014	-7.4E+03	<.0001	29.0	<.0001
82	-2.5445	0.5486	<.0001	0.01346	0.1545	-1.0E+04	<.0001	37.1	<.0001
151	-2.7581	0.7845	<.0001	0.0169	<.0001	-6.8E+03	<.0001	24.9	<.0001
135+144+147+124	-2.9481	0.763	<.0001	0.01722	0.0002	-7.3E+03	<.0001	27.1	<.0001
149+123+107	-2.9549	0.6347	<.0001	0.02115	0.001	-7.2E+03	<.0001	27.3	<.0001
118	-2.9310	0.7336	<.0001	0.0183	0.001	-7.6E+03	<.0001	29.0	<.0001
146	-3.1211	0.8221	<.0001	0.01861	<.0001	-9.0E+03	<.0001	32.2	<.0001
153+132	-3.1735	0.7959	<.0001	0.01926	0.0001	-8.7E+03	<.0001	32.6	<.0001
105	-3.0664	0.6901	<.0001	0.01988	0.0048	-9.5E+03	<.0001	34.3	<.0001
141	-3.2211	0.7729	<.0001	0.0215	<.0001	-8.5E+03	<.0001	30.3	<.0001
137+176+130	-3.5511	0.4908	<.0001	0.02398	0.0218	-8.6E+03	<.0001	28.9	0.0002
163+138	-3.2311	0.8156	<.0001	0.02246	<.0001	-9.8E+03	<.0001	36.4	<.0001
158	-3.2211	0.8238	<.0001	0.02154	<.0001	-9.6E+03	<.0001	33.7	<.0001
178+129	-3.3710	0.7139	<.0001	0.01638	0.0113	-9.5E+03	<.0001	33.4	<.0001
187+182	-3.5199	0.8864	<.0001	0.01521	<.0001	-9.8E+03	<.0001	35.0	<.0001
183	-3.4610	0.8672	<.0001	0.02014	<.0001	-1.0E+04	<.0001	35.2	<.0001
128	-3.4692	0.637	<.0001	0.02127	0.0205	-1.1E+04	<.0001	38.6	<.0001
185	-3.5010	0.8887	<.0001	0.01881	<.0001	-1.0E+04	<.0001	33.1	<.0001

CONGENER GROUP	log p	R <sup>2</sup>	p-value	a <sub>1</sub> (RH)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
174	-3.7510	0.8496	<.0001	0.01899	<.0001	-1.0E+04	<.0001	35.1	<.0001
177	-3.6110	0.8358	<.0001	0.01829	0.0002	-1.0E+04	<.0001	35.7	<.0001
202+171+156	-3.2885	0.8733	<.0001	0.01573	0.0012	-1.2E+04	<.0001	42.5	<.0001
180	-3.8895	0.8762	<.0001	0.02143	<.0001	-1.3E+04	<.0001	44.4	<.0001
199	-4.2373	0.9059	<.0001	0.0166	<.0001	-1.2E+04	<.0001	40.3	<.0001
170+190	-4.0750	0.877	<.0001	0.02067	0.0001	-1.3E+04	<.0001	44.6	<.0001
201	-3.7685	0.9089	<.0001	0.01582	0.0009	-1.4E+04	<.0001	50.7	<.0001
203+196	-4.2073	0.9218	<.0001	0.01641	0.0002	-1.4E+04	<.0001	50.6	<.0001
195+208	-4.3573	0.5956	<.0001	0.0071	0.4943	-1.3E+04	<.0001	44.9	<.0001
194	-4.6975	0.9036	<.0001	0.01599	0.001	-1.4E+04	<.0001	47.0	<.0001
206	-4.9736	0.7813	<.0001	0.01301	0.0573	-1.3E+04	<.0001	45.5	<.0001

Table I-4f. Summary of multiple linear regressions including temperature (1/T) and solar irradiance (SR).

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (SR)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
CONGENER GROOT	log p	I N	p-varue	a <sub>1</sub> (SK)	p-value	a <sub>2</sub> (1/1)	p-value	тистесрі	p-varue
18	-1.1147	0.3946	<.0001	-2.2926	0.0712	-7.2E+03	<.0001	30.1	<.0001
17+15	-1.1547	0.4071	<.0001	-1.8548	0.1152	-6.6E+03	<.0001	27.4	<.0001
16+32	-1.2747	0.4084	<.0001	-1.6097	0.2133	-7.3E+03	<.0001	30.4	<.0001
31	-1.4674	0.5239	<.0001	-2.2733	0.0516	-8.4E+03	<.0001	34.6	<.0001
28	-1.4745	0.478	<.0001	-1.7139	0.0904	-6.7E+03	<.0001	28.0	<.0001
21+33+53	-1.5645	0.486	<.0001	-1.2028	0.3066	-7.7E+03	<.0001	31.3	<.0001
22	-1.5945	0.401	<.0001	-1.2234	0.3974	-7.9E+03	<.0001	31.6	<.0001
45	-1.6554	0.5152	<.0001	-1.6769	0.1054	-7.3E+03	<.0001	28.1	<.0001
46	-1.7000	0.5864	0.0001	-1.8711	0.2738	-1.1E+04	<.0001	40.2	<.0001
52+43	-1.8011	0.623	<.0001	-1.8727	0.033	-7.6E+03	<.0001	32.2	<.0001
49	-1.7813	0.6268	<.0001	-2.4123	0.0063	-7.8E+03	<.0001	31.6	<.0001

CONGENER GROUP	log p	R <sup>2</sup>	p-value	a <sub>1</sub> (SR)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
47+48	-1.8213	0.5673	<.0001	-2.5836	0.0087	-7.7E+03	<.0001	31.0	<.0001
44	-1.9013	0.6087	<.0001	-1.6641	0.0749	-7.8E+03	<.0001	32.3	<.0001
37+42	-1.9049	0.5712	<.0001	-1.5612	0.1422	-8.2E+03	<.0001	32.6	<.0001
41+71	-1.9713	0.5527	<.0001	-2.2171	0.0593	-8.9E+03	<.0001	35.1	<.0001
64	-1.8913	0.5647	<.0001	-1.4487	0.147	-7.6E+03	<.0001	30.2	<.0001
40	-2.0122	0.545	<.0001	-0.7547	0.572	-9.5E+03	<.0001	35.0	<.0001
74	-2.1447	0.655	<.0001	-2.057	0.0204	-8.2E+03	<.0001	32.7	<.0001
70+76	-2.2691	0.67	<.0001	-1.7822	0.037	-8.2E+03	<.0001	33.2	<.0001
66+95	-2.2140	0.6651	<.0001	-1.4767	0.0801	-7.9E+03	<.0001	33.4	<.0001
91	-2.3217	0.6824	<.0001	-0.9998	0.1808	-7.2E+03	<.0001	28.4	<.0001
56+60+89	-2.3747	0.6079	<.0001	-0.9779	0.3238	-8.1E+03	<.0001	32.2	<.0001
92+84	-2.4145	0.5503	<.0001	-0.8251	0.4404	-7.7E+03	<.0001	31.4	<.0001
101	-2.4777	0.6923	<.0001	-1.7086	0.0344	-8.1E+03	<.0001	33.4	<.0001
99	-2.5314	0.6958	<.0001	-2.0021	0.0194	-8.7E+03	<.0001	34.5	<.0001
83	-2.5745	0.7196	<.0001	-1.8312	0.027	-8.6E+03	<.0001	32.4	<.0001
97	-2.6145	0.605	<.0001	-1.9487	0.0393	-7.9E+03	<.0001	31.3	<.0001
87+81	-2.6487	0.6119	<.0001	-0.8546	0.3895	-8.2E+03	<.0001	32.7	<.0001
85+136	-2.6345	0.6025	<.0001	-2.5078	0.0309	-9.7E+03	<.0001	37.6	<.0001
110+77	-2.7445	0.5461	<.0001	-1.6622	0.1599	-8.7E+03	<.0001	35.4	<.0001
82	-2.5445	0.5259	<.0001	0.33639	0.8372	-1.0E+04	<.0001	37.8	<.0001
151	-2.7581	0.725	<.0001	-1.9259	0.011	-8.2E+03	<.0001	31.4	<.0001
135+144+147+124	-2.9481	0.6995	<.0001	-1.6808	0.0472	-8.6E+03	<.0001	33.1	<.0001
149+123+107	-2.9549	0.5702	<.0001	-2.3648	0.0381	-9.0E+03	<.0001	35.4	<.0001
118	-2.9310	0.6332	<.0001	-1.1962	0.2366	-8.8E+03	<.0001	34.6	<.0001
146	-3.1211	0.7597	<.0001	-1.5382	0.0793	-1.0E+04	<.0001	38.0	<.0001
153+132	-3.1735	0.7312	<.0001	-1.718	0.062	-1.0E+04	<.0001	38.9	<.0001
105	-3.0664	0.6243	<.0001	-0.7519	0.5477	-1.0E+04	<.0001	38.7	<.0001
141	-3.2211	0.6981	<.0001	-2.028	0.0413	-1.0E+04	<.0001	37.6	<.0001
137+176+130	-3.5511	0.4184	0.0003	-2.1068	0.2466	-1.0E+04	<.0001	37.6	0.0001
163+138	-3.2311	0.7487	<.0001	-2.1119	0.0355	-1.1E+04	<.0001	44.1	<.0001
158	-3.2211	0.7666	<.0001	-2.2954	0.0155	-1.1E+04	<.0001	41.7	<.0001
178+129	-3.3710	0.6748	<.0001	-1.3192	0.2431	-1.1E+04	<.0001	38.7	<.0001
187+182	-3.5199	0.8476	<.0001	-1.4814	0.0322	-1.1E+04	<.0001	40.2	<.0001
183	-3.4610	0.8207	<.0001	-2.2109	0.0093	-1.2E+04	<.0001	42.8	<.0001

CONGENER GROUP	log p	$\mathbb{R}^2$	p-value	a <sub>1</sub> (SR)	p-value	a <sub>2</sub> (1/T)	p-value	intercept	p-value
128	-3.4692	0.5842	<.0001	-0.0043	0.9979	-1.2E+04	<.0001	41.4	<.0001
185	-3.5010	0.8494	<.0001	-2.1928	0.004	-1.2E+04	<.0001	40.5	<.0001
174	-3.7510	0.8071	<.0001	-2.0519	0.0187	-1.2E+04	<.0001	42.2	<.0001
177	-3.6110	0.7952	<.0001	-1.9703	0.0272	-1.2E+04	<.0001	42.4	<.0001
202+171+156	-3.2885	0.8472	<.0001	-1.4925	0.0851	-1.3E+04	<.0001	47.9	<.0001
180	-3.8895	0.8447	<.0001	-2.5208	0.0093	-1.5E+04	<.0001	52.9	<.0001
199	-4.2373	0.8767	<.0001	-1.6187	0.0363	-1.3E+04	<.0001	46.1	<.0001
170+190	-4.0750	0.8419	<.0001	-2.0421	0.0389	-1.5E+04	<.0001	51.9	<.0001
201	-3.7685	0.8898	<.0001	-1.5471	0.0704	-1.5E+04	<.0001	56.3	<.0001
203+196	-4.2073	0.9005	<.0001	-1.5613	0.0538	-1.5E+04	<.0001	56.2	<.0001
195+208	-4.3573	0.6045	<.0001	2.06661	0.2419	-1.2E+04	<.0001	40.7	<.0001
194	-4.6975	0.8824	<.0001	-1.4679	0.0911	-1.5E+04	<.0001	52.4	<.0001
206	-4.9736	0.7669	<.0001	-1.2438	0.2941	-1.4E+04	<.0001	49.9	<.0001

Table I-5. Summary of enthalpies; SAS calculated ( $\Delta H_{SA}$ ) and literature reported values ( $\Delta H_{SA}$  represents the enthalpy of change between surface and air;  $\Delta H_{VAP}$  represents the enthalpy of change between plant surface and air;  $\Delta H_{VAP}$  represents enthalpy of vaporization).

Congener Group	$\Delta H_{SA}$ (This study)	$\Delta H_{VAP}^{a}$	$\Delta H_{VAP}^{b}$	$\Delta H_{VAP}^{^c}$	$\Delta H_{VAP}^{d}$	$\Delta H_{VAP}^{\ \ h}$	$\Delta H_{VAP}^{i}$	$\Delta H_{VAP}^{j}$	$\Delta H_{VAP}^{k}$	$\Delta \mathbf{H}_{\mathbf{SA}}^f$	$\Delta H_{SA}{}^g$	$\Delta \mathrm{H_{PA}}^e$
16+32	50.22	75.35		38.26		79.9		80.7	77.4			64.5
17+15	43.37	75.35		38.18		81.6	73.45	83.6	77.8			
18	45.37	75.35		32.93	65.0	81.5	76.25	81.2	77.6	57.0	23.5	70.6
21+33+53	54.76	78.03				88.6		86.6	78.6			
22	56.73	78.03		34.77	70.0	81.0	76.45	84.8	78.2	49.0	41.7	
28	44.05	78.03	67.98	34.16		89.3	79.44	86.0	78.4			
31	55.71	77.70	93.44336		70.0	91.2	83.57	84.9	78.0	52.0	27.7	82.4

Congener Group	$\Delta H_{SA}$ (This study)	$\Delta H_{VAP}^{a}$	$\Delta \mathrm{H}_{\mathrm{VAP}}^{}b}$	$\Delta H_{VAP}^{c}$	$\Delta \mathbf{H}_{\mathrm{VAP}}^{}d}$	$\Delta \mathrm{H}_{\mathrm{VAP}}^{}h}$	$\Delta \mathbf{H_{VAP}}^i$	$\Delta \mathbf{H}_{\mathbf{VAP}}{}^{j}$	$\Delta \mathbf{H}_{\mathrm{VAP}}^{}^{}}}$	$\Delta \mathbf{H}_{\mathbf{S}\mathbf{A}}^f$	$\Delta H_{SA}{}^g$	$\Delta H_{PA}^{e}$
37+42	57.97	81.23				91.1		88.5	78.8			
40	72.18	81.78				94.7		86.4	82.3	27.0		
41+71	60.19	80.98				90.5		91.3	82.9	44.0	53.8	
44	54.21	80.98			76.0	84.4	83.26	86.5	82.0	37.0	58.9	83.7
45	49.35	78.83			44.0	80.5		88.9	82.4	46.0		
46	78.85	78.83			69.0	85.9		84.8	81.8			
47+48	48.53	80.98	76.21			90.3		90.0	83.0	45.0	50.8	
49	49.73	80.98	72.38	36.59	72.0	87.0		87.4	82.2	37.0	47.7	
52+43	51.43	80.81	71.81	42.77	77.0	84.5	81.28	86.8	82.0	46.0	59.4	86.6
56+60+89	59.53	83.91			67.0	99.7		92.5	83.1	42.0	72.0	
64	53.57	80.98			81.0	91.7		92.5	83.6			
66+95	55.79	83.28			69.0	93.0	78.0	93.8	83.8	48.0		
70+76	56.20	84.85	64.53	31.98	70.0	96.1	83.41	93.4	83.8	51.0	70.9	
74	55.44	83.91			67.0	94.4		94.7	84.1	48.0		
82	85.28	86.59				92.5		96.3	88.1	53.0		
83	59.71	86.59			60.0	91.7		95.0	87.6	51.0		
85+136	65.40	86.59			28.0	100.2		97.3	87.9			
87+81	60.78	87.35			75.0	99.6		94.8	87.6	65.0	73.3	
91	52.13	84.23			66.0	89.2		94.9	87.6	57.0	60.3	
92+84	57.23	86.59			67.0	97.0		95.1	87.4			
97	53.46	86.59		24.53	73.0	96.3		96.5	87.8	66.0	72.9	
99	59.26	86.80	80.04	-19.15		102.3		97.1	88.3	66.0	71.3	
101	56.00	86.44	65.49	25.74	78.0	95.6	87.11	96.9	87.8		70.5	
105	79.21	91.11				106.7		101.0	88.7	61.0	97.8	
110+77	61.45	86.59		25.16	98.0	97.6	89.58	96.9	87.8	67.0	79.7	107.2
118	64.08	89.31				107.6	87.03	102.0	89.2	72.0	85.5	
128	94.57	93.46				106.0		103.0	93.1			
135+144+147+124	60.36	89.63		72.19	79.0	97.0		98.5	91.8	75.0		

Congener Group	$\Delta H_{SA}$ (This study)	$\Delta H_{VAP}^{a}$	$\Delta H_{VAP}^{b}$	$\Delta H_{VAP}^{c}$	$\Delta \mathbf{H}_{\mathrm{VAP}}^{}^{}}}$	$\Delta H_{VAP}^{\ \ h}$	$\Delta H_{VAP}^{i}$	$\Delta H_{VAP}^{j}$	$\Delta H_{VAP}^{k}$	$\Delta \mathbf{H}_{\mathbf{S}\mathbf{A}}^{f}$	$\Delta H_{SA}{}^g$	$\Delta H_{PA}^{e}$
137+176+130	73.39	92.22			66.0	105.3		105.0	93.7			
141	70.77	92.22			83.0	106.2		104.0	93.2	73.0		
146	74.43	92.22				106.8		102.0	93.0			
149+123+107	59.87	89.79		65.45	74.0	97.1	90.01	98.7	92.3	77.0	47.0	108.9
151	55.71	89.63	60.89	76.23	65.0	92.0		103.0	93.0	80.0	63.7	
153+132	71.94	91.43	71.61	60.36	67.0	103.4	99.38	102.0	93.0	90.0	77.1	116.7
158	79.88	92.22				110.7		107.0	94.2			123.3
163+138	81.76	92.22			76.0	106.7		107.0	94.2			
170+190	109.51	98.40		106.37	65.0	116.0	97.38	110.0	98.7			
174	83.77	95.01		87.89	57.0	103.3	95.56	107.0	97.7	94.0		
177	85.36	95.01			58.0	102.9		108.0	97.7	73.0	99.2	
178+129	79.02	95.01			44.0	105.5		108.0	97.7			
180	105.98	96.55		79.48	47.0	111.9	99.37	110.0	97.9	112.0	83.2	128.8
183	84.56	95.01			51.0	113.1		109.0	97.9	99.0		
185	83.37	95.01			61.0	99.6		111.0	98.7			
187+182	79.64	94.04	76.02		49.0	106.0		108.0	97.7	106.0		117.2
194	116.02	103.44				119.6		115.0	103.4			
195+208	106.51	100.41				117.3		118	103.6			
199	98.83	100.41				112.1		113	102.7			
201	118.14	92.89			47.0	109.2		111.0	101.9			
202+171+156	98.86	92.89				104.7		109.0	102.0			108.8
203+196	118.04	100.41			31.0	110.7		117.0	103.6			137.5
206	108.99	105.81				119.0		123.0	108.3			
$\Sigma$ PCBs (incl. 8+5)	53 13											

 $\Sigma$ PCBs (incl. 8+5) 53.13

 $\sum$  PCBs (w/o 8+5) 55.84

<sup>a</sup>Falconer & Bidleman, 1994. <sup>b</sup>Hoff et al <sup>c</sup>Hornbuckle & Eisenreich, 1996. <sup>d</sup>Panshin & Hites, 1994. <sup>e</sup>Komp & McCachlan, 1997. <sup>f</sup>Currado and Harrad, 2000. <sup>g</sup>Harrad and Mao, 2004. <sup>h</sup>Puri et al, 2002. <sup>i</sup>Najakoh et al, 2006. <sup>ccc</sup> et al, 2007; using Puri et al, 2001. <sup>k</sup>Padmanabhan et al, 2007; using Nak et al, 2006.

Table I-6. Multiple regression analysis including WS, RH, SR and 1/T.

CONGENER	_ 2							_	l	_		
GROUP	R <sup>2</sup>	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (RH)	p-value	a <sub>3</sub> (SR)	p-value	a <sub>4</sub> (1/T)	p-value	intercept	p-value
18	0.549	<.0001	-0.194	0.024	0.016	0.104	-0.131	0.936	-5.10E+03	0.0016	22.0	0.0007
17+15	0.571	<.0001	-0.209	0.014	0.012	0.209	-0.175	0.906	-4.78E+03	0.0015	20.5	0.0007
16+32	0.596	<.0001	-0.218	0.011	0.020	0.052	0.946	0.554	-4.91E+03	0.002	20.9	0.001
31	0.681	<.0001	-0.197	0.009	0.018	0.043	0.071	0.960	-6.19E+03	<.0001	25.9	<.0001
28	0.687	<.0001	-0.172	0.006	0.019	0.010	0.777	0.504	-4.39E+03	0.0002	18.9	<.0001
21+33+53	0.691	<.0001	-0.182	0.035	0.018	0.051	0.998	0.477	-5.44E+03	0.0008	22.5	0.0005
22	0.572	<.0001	-0.228	0.019	0.021	0.062	1.576	0.392	-5.24E+03	0.0037	21.2	0.0031
45	0.699	<.0001	-0.174	0.008	0.019	0.014	0.779	0.521	-5.03E+03	<.0001	19.1	0.0001
46	0.714	<.0001	-0.143	0.267	0.025	0.103	1.879	0.435	-6.72E+03	0.0172	23.3	0.0323
52+43	0.737	<.0001	-0.141	0.014	0.013	0.058	-0.199	0.854	-6.06E+03	<.0001	26.0	<.0001
49	0.750	<.0001	-0.145	0.009	0.013	0.043	-0.678	0.515	-6.14E+03	<.0001	25.2	<.0001
47+48	0.701	<.0001	-0.152	0.016	0.015	0.045	-0.637	0.591	-5.93E+03	<.0001	23.8	<.0001
44	0.744	<.0001	-0.152	0.011	0.016	0.025	0.403	0.719	-5.95E+03	<.0001	24.7	<.0001
37+42	0.680	<.0001	-0.131	0.067	0.018	0.039	0.718	0.600	-6.23E+03	<.0001	24.4	<.0001
41+71	0.697	<.0001	-0.193	0.011	0.018	0.043	0.157	0.913	-6.68E+03	<.0001	26.3	<.0001
64	0.715	<.0001	-0.152	0.018	0.019	0.015	0.943	0.434	-5.50E+03	<.0001	21.5	<.0001
40	0.651	<.0001	-0.179	0.100	0.014	0.249	0.899	0.619	-7.71E+03	0.0003	28.3	0.0007
74	0.757	<.0001	-0.127	0.027	0.014	0.035	-0.189	0.861	-6.55E+03	<.0001	25.8	<.0001
70+76	0.771	<.0001	-0.121	0.029	0.015	0.026	0.108	0.918	-6.47E+03	<.0001	26.3	<.0001
66+95	0.771	<.0001	-0.123	0.024	0.015	0.024	0.429	0.678	-6.21E+03	<.0001	26.4	<.0001
91	0.794	<.0001	-0.106	0.025	0.015	0.009	0.913	0.312	-5.51E+03	<.0001	21.6	<.0001
56+60+89	0.704	<.0001	-0.137	0.043	0.015	0.071	0.909	0.482	-6.33E+03	<.0001	25.3	<.0001
92+84	0.687	<.0001	-0.171	0.017	0.017	0.047	1.358	0.314	-5.65E+03	<.0001	23.3	<.0001
101	0.779	<.0001	-0.104	0.049	0.014	0.029	0.074	0.941	-6.52E+03	<.0001	27.0	<.0001
99	0.776	<.0001	-0.101	0.071	0.015	0.028	-0.107	0.920	-7.02E+03	<.0001	27.7	<.0001
83	0.786	<.0001	-0.036	0.529	0.018	0.008	0.381	0.722	-6.90E+03	<.0001	24.8	<.0001
97	0.713	<.0001	-0.116	0.061	0.017	0.027	0.169	0.886	-6.09E+03	<.0001	23.7	<.0001
87+81	0.749	<.0001	-0.136	0.068	0.015	0.058	1.009	0.413	-6.31E+03	<.0001	25.4	<.0001
85+136	0.653	<.0001	-0.069	0.401	0.017	0.092	-0.414	0.794	-8.01E+03	<.0001	30.3	<.0001

CONGENER GROUP	$\mathbb{R}^2$	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (RH)	p-value	a <sub>3</sub> (SR)	p-value	a <sub>4</sub> (1/T)	p-value	intercept	p-value
110+77	0.676	<.0001	-0.151	0.054	0.022	0.023	1.081	0.469	-6.32E+03	<.0001	25.6	<.0001
82	0.607	<.0001	-0.176	0.133	0.022	0.118	3.148	0.166	-7.82E+03	0.0006	27.6	0.0018
151	0.804	<.0001	-0.092	0.059	0.014	0.020	-0.181	0.845	-6.68E+03	<.0001	25.2	<.0001
135+144+147+124	0.787	<.0001	-0.104	0.059	0.016	0.018	0.336	0.749	-6.86E+03	<.0001	25.9	<.0001
149+123+107	0.680	<.0001	-0.172	0.024	0.015	0.099	-0.410	0.776	-7.10E+03	<.0001	28.1	<.0001
118	0.781	<.0001	-0.127	0.079	0.019	0.020	1.050	0.382	-6.63E+03	<.0001	25.9	<.0001
146	0.845	<.0001	-0.101	0.063	0.020	0.003	0.981	0.348	-8.10E+03	<.0001	29.1	<.0001
153+132	0.823	<.0001	-0.121	0.038	0.019	0.008	0.684	0.535	-7.95E+03	<.0001	30.3	<.0001
105	0.739	<.0001	-0.123	0.132	0.028	0.007	2.695	0.091	-7.36E+03	<.0001	26.3	<.0001
141	0.785	<.0001	-0.067	0.290	0.023	0.004	0.863	0.485	-7.78E+03	<.0001	27.7	<.0001
137+176+130	0.518	0.0003	-0.163	0.280	0.020	0.215	0.355	0.889	-8.09E+03	0.0046	27.9	0.0122
163+138	0.836	<.0001	-0.115	0.066	0.022	0.004	0.701	0.557	-9.09E+03	<.0001	34.2	<.0001
158	0.839	<.0001	-0.104	0.083	0.019	0.008	0.157	0.891	-9.27E+03	<.0001	33.0	<.0001
178+129	0.739	<.0001	-0.131	0.100	0.016	0.105	0.668	0.659	-8.78E+03	<.0001	31.3	<.0001
187+182	0.905	<.0001	-0.104	0.015	0.013	0.009	0.215	0.787	-9.39E+03	<.0001	34.3	<.0001
183	0.878	<.0001	-0.094	0.076	0.018	0.006	0.032	0.975	-9.96E+03	<.0001	34.9	<.0001
128	0.705	<.0001	-0.143	0.180	0.036	0.007	4.444	0.035	-7.94E+03	0.0002	25.5	0.0018
185	0.896	<.0001	-0.077	0.106	0.016	0.006	-0.176	0.846	-1.00E+04	<.0001	33.4	<.0001
174	0.860	<.0001	-0.087	0.122	0.017	0.014	0.101	0.925	-9.84E+03	<.0001	34.6	<.0001
177	0.848	<.0001	-0.095	0.105	0.016	0.025	0.023	0.983	-1.02E+04	<.0001	35.5	<.0001
202+171+156	0.893	<.0001	-0.140	0.015	0.013	0.058	0.178	0.868	-1.15E+04	<.0001	41.6	<.0001
180	0.884	<.0001	-0.102	0.107	0.017	0.023	-0.316	0.793	-1.27E+04	<.0001	45.1	<.0001
199	0.922	<.0001	-0.124	0.010	0.014	0.014	0.211	0.815	-1.15E+04	<.0001	39.4	<.0001
170+190	0.883	<.0001	-0.072	0.268	0.021	0.008	0.610	0.626	-1.26E+04	<.0001	42.7	<.0001
201	0.922	<.0001	-0.133	0.019	0.013	0.055	0.124	0.907	-1.39E+04	<.0001	50.1	<.0001
203+196	0.933	<.0001	-0.117	0.024	0.015	0.019	0.317	0.746	-1.38E+04	<.0001	49.4	<.0001
195+208	0.652	<.0001	-0.048	0.708	0.030	0.058	5.699	0.026	-9.01E+03	0.0003	28.3	0.0034
194	0.912	<.0001	-0.099	0.089	0.015	0.029	0.489	0.660	-1.35E+04	<.0001	45.4	<.0001
206	0.789	<.0001	-0.098	0.262	0.011	0.280	0.186	0.912	-1.30E+04	<.0001	44.8	<.0001

Table I-7. Multiple regression analysis including WS, RH and 1/T.

CONGENER GROUP	$\mathbb{R}^2$	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (RH)	p-value	a <sub>3</sub> (1/T)	p-value	intercept	p-value
18	0.549	<.0001	-0.193	0.019	0.017	0.014	-5.01E+03	<.0001	21.6	<.0001
17+15	0.571	<.0001	-0.206	0.015	0.017	0.014	-4.66E+03	<.0001	20.0	<.0001
16+32	0.592	<.0001	-0.230	0.011	0.015	0.031	-5.52E+03	<.0001	23.6	<.0001
31	0.681	<.0001	-0.198	0.006	0.013	0.020	-6.24E+03	<.0001	26.1	<.0001
28	0.683	<.0001	-0.198	0.000	0.016	0.004	-4.90E+03	<.0001	21.1	<.0001
21+33+53	0.685	<.0001	-0.182	0.003	0.010	0.002	-4.90E+03 -6.15E+03	<.0001	25.6	<.0001
21+35+35	0.564	<.0001	-0.248	0.012	0.013	0.066	-6.27E+03	<.0001	25.7	<.0001
45	0.696	<.0001	-0.184	0.003	0.014	0.004	-5.54E+03	<.0001	21.4	<.0001
46	0.704	<.0001	-0.170	0.169	0.016	0.004	-3.34E+03 -8.23E+03	0.0001	29.7	<.0001
52+43	0.704	<.0001	-0.170	0.109	0.010	0.094	-5.93E+03	<.0001	25.4	<.0001
49	0.730	<.0001	-0.136	0.012	0.014	0.004	-5.70E+03	<.0001	23.4	<.0001
47+48	0.698	<.0001	-0.144	0.017	0.018	0.001	-5.51E+03	<.0001	22.0	<.0001
44	0.743	<.0001	-0.157	0.006	0.014	0.004	-6.21E+03	<.0001	25.8	<.0001
37+42	0.678	<.0001	-0.140	0.042	0.014	0.013	-6.70E+03	<.0001	26.4	<.0001
41+71	0.697	<.0001	-0.195	0.008	0.013	0.005	-6.78E+03	<.0001	26.7	<.0001
64	0.711	<.0001	-0.164	0.008	0.017	0.006	-6.11E+03	<.0001	24.2	<.0001
40	0.648	<.0001	-0.199	0.050	0.009	0.235	-8.34E+03	<.0002	31.1	<.0001
74	0.757	<.0001	-0.124	0.024	0.015	0.001	-6.43E+03	<.0001	25.3	<.0001
70+76	0.771	<.0001	-0.122	0.021	0.013	0.002	-6.54E+03	<.0001	26.6	<.0001
66+95	0.770	<.0001	-0.129	0.015	0.013	0.004	-6.49E+03	<.0001	27.7	<.0001
91	0.788	<.0001	-0.118	0.011	0.011	0.006	-6.10E+03	<.0001	24.2	<.0001
56+60+89	0.701	<.0001	-0.149	0.024	0.010	0.056	-6.92E+03	<.0001	27.9	<.0001
92+84	0.678	<.0001	-0.188	0.007	0.011	0.063	-6.54E+03	<.0001	27.2	<.0001
101	0.779	<.0001	-0.105	0.038	0.014	0.002	-6.57E+03	<.0001	27.2	<.0001
99	0.776	<.0001	-0.100	0.063	0.015	0.001	-6.95E+03	<.0001	27.4	<.0001
83	0.785	<.0001	-0.041	0.438	0.016	0.001	-7.15E+03	<.0001	25.9	<.0001
97	0.713	<.0001	-0.119	0.047	0.016	0.002	-6.20E+03	<.0001	24.2	<.0001
87+81	0.743	<.0001	-0.158	0.025	0.011	0.050	-7.03E+03	<.0001	28.5	<.0001
85+136	0.653	<.0001	-0.064	0.418	0.019	0.007	-7.74E+03	<.0001	29.1	<.0001

CONGENER GROUP	$\mathbb{R}^2$	p-value	a <sub>1</sub> (WS)	p-value	a <sub>2</sub> (RH)	p-value	a <sub>3</sub> (1/T)	p-value	intercept	p-value
110+77	0.671	<.0001	-0.165	0.030	0.017	0.010	-7.03E+03	<.0001	28.7	<.0001
82	0.587	<.0001	-0.216	0.062	0.008	0.417	-9.87E+03	<.0001	36.7	<.0001
151	0.804	<.0001	-0.090	0.054	0.015	0.001	-6.56E+03	<.0001	24.7	<.0001
135+144+147+124	0.787	<.0001	-0.108	0.041	0.014	0.002	-7.07E+03	<.0001	26.9	<.0001
149+123+107	0.680	<.0001	-0.167	0.023	0.017	0.007	-6.83E+03	<.0001	26.9	<.0001
118	0.775	<.0001	-0.150	0.028	0.014	0.011	-7.37E+03	<.0001	29.1	<.0001
146	0.842	<.0001	-0.114	0.032	0.016	0.001	-8.74E+03	<.0001	31.9	<.0001
153+132	0.822	<.0001	-0.129	0.021	0.016	0.001	-8.39E+03	<.0001	32.3	<.0001
105	0.719	<.0001	-0.159	0.053	0.015	0.028	-9.14E+03	<.0001	34.1	<.0001
141	0.782	<.0001	-0.078	0.203	0.019	0.001	-8.34E+03	<.0001	30.1	<.0001
137+176+130	0.517	<.0001	-0.171	0.218	0.019	0.088	-8.34E+03	0.0001	29.0	0.0002
163+138	0.834	<.0001	-0.124	0.040	0.019	0.000	-9.54E+03	<.0001	36.2	<.0001
158	0.838	<.0001	-0.106	0.065	0.019	0.000	-9.37E+03	<.0001	33.5	<.0001
178+129	0.738	<.0001	-0.139	0.068	0.013	0.055	-9.22E+03	<.0001	33.2	<.0001
187+182	0.905	<.0001	-0.107	0.009	0.012	0.001	-9.53E+03	<.0001	34.9	<.0001
183	0.878	<.0001	-0.094	0.063	0.018	0.000	-9.98E+03	<.0001	35.0	<.0001
128	0.668	<.0001	-0.201	0.065	0.016	0.090	-1.09E+04	<.0001	38.4	<.0001
185	0.896	<.0001	-0.074	0.101	0.017	<.0001	-9.90E+03	<.0001	32.9	<.0001
174	0.860	<.0001	-0.089	0.101	0.017	0.001	-9.91E+03	<.0001	34.9	<.0001
177	0.848	<.0001	-0.095	0.089	0.016	0.002	-1.02E+04	<.0001	35.6	<.0001
202+171+156	0.893	<.0001	-0.142	0.010	0.012	0.010	-1.16E+04	<.0001	42.2	<.0001
180	0.884	<.0001	-0.098	0.105	0.019	0.001	-1.25E+04	<.0001	44.2	<.0001
199	0.922	<.0001	-0.127	0.006	0.013	0.001	-1.17E+04	<.0001	40.0	<.0001
170+190	0.882	<.0001	-0.079	0.202	0.019	0.001	-1.30E+04	<.0001	44.4	<.0001
201	0.922	<.0001	-0.135	0.013	0.012	0.008	-1.40E+04	<.0001	50.4	<.0001
203+196	0.933	<.0001	-0.121	0.015	0.013	0.002	-1.40E+04	<.0001	50.3	<.0001
195+208	0.604	<.0001	-0.120	0.357	0.004	0.719	-1.27E+04	<.0001	44.7	<.0001
194	0.912	<.0001	-0.105	0.060	0.013	0.006	-1.38E+04	<.0001	46.8	<.0001
206	0.789	<.0001	-0.100	0.230	0.010	0.141	-1.31E+04	<.0001	45.4	<.0001