LBL-30149
UC-407

Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

ENERGY & ENVIRONMENT DIVISION

Potential for Ion-Induced Nucleation of Volatile Organic Compounds by Radon Decay in Indoor Environments

J.M. Daisey

November 1991

ENERGY & ENVIRONMENT DIVISION

| LOAN COPY |
|Circulates |
|for 4 weeks| Bldg. 50 Library.
LBL-30149
Copy 2

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

LBL-30149

POTENTIAL FOR ION-INDUCED NUCLEATION OF VOLATILE ORGANIC COMPOUNDS BY RADON DECAY IN INDOOR ENVIRONMENTS

by

Joan M. Daisey
Indoor Environment Program
Energy and Environment Division
Lawrence Berkeley Laboratory
Berkeley, CA 94720

November 4, 1991

This research was supported the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division, U.S. Department of Energy, under Contract No. DE-AC03-76SF00098.

ABSTRACT

There is considerable interest in the "unattached" fraction of radon progeny in indoor air because of its significance to the estimation of the risks of radon exposure. Because of its high mobility in air, the unattached fraction is more efficiently deposited in the respiratory tract. Variation in the diameter of the "unattached" fraction and in its diffusion coefficient can be due to clustering of other atmospheric species around the $^{218}PoO_2^+$ ion.

The purpose of this study was to investigate the potential for the formation of clusters of vapor phase organic compounds, found in indoor air, around the $^{218}PoO_2^+$ ion and to determine which were most likely to form clusters. A secondary purpose was to provide a compilation of measurements of indoor organic compounds for future experiments and theoretical calculations by the radon research community.

The classical charged liquid droplet theory (Thomson equation) was used to estimate the Gibbs free energy of ion-induced nucleation and to provide an indication of the indoor organic compounds most likely to undergo ion-induced nucleation. Forty-four volatile and semi-volatile organic compounds out of the more than 300 which have been reported in indoor air were investigated. Water vapor was included for comparison.

The results indicate that there is a potential for the formation of clusters of organic compounds around the $^{218}PoO_2^+$ ion. The compounds with the greatest potential for cluster formation are the volatile oxidized hydrocarbons (e.g., n-butanol, phenol, hexanal, nonanal, benzaldehyde, the ketones and the acetates) and the semi-volatile organic compounds (pentachlorophenol, nicotine, chlordane, chlorpyrifos). The theory estimates an average of one VOC molecule per ion and cluster diameters of 0.6 to 1.0 nm. Although the estimated diameters are consistent with the measured diameters for the "unattached" fraction, the state of experimental and theoretical knowledge in this area is not sufficiently developed to judge the validity of these predictions. Experimental investigations of cluster sizes for the VOC and SVOC most likely to form clusters are recommended.

The results of this work also have implications for experimental determinations of the diffusion coefficient of the $^{218}PoO_2^+$ ion. Since ambient air always has VOC and SVOC present, it should be purified to remove these compounds in any determinations of diffusion coefficients. Furthermore, sources of VOC and SVOC contaminants in the experimental apparatus should be considered. For example Tygon tubing, which is widely used in laboratories, is a source of phthalate esters (SVOC) which have some potential for cluster formation.

INTRODUCTION

There is considerable interest in the "unattached" fraction of radon progeny in indoor air because of its significance to the estimation of the risks of radon exposure (Strong, 1988; Reineking, et al., 1988). The "unattached" fraction appears to be about 0.5 to 3 nm in diameter (Chu, et al., 1987; Strong, 1988; Reineking, et al., 1988; Tu, et al., 1991). Because of its high mobility in air, the unattached fraction is more efficiently deposited in the respiratory tract. Typically, most of the unattached fraction is Po-218 (Hopke, 1987). Variations in the diameter of this fraction and in its diffusion coefficient can be due to clustering of other atmospheric species around the $^{218}PoO_2^+$ ion (Raabe, 1968; Porstendorfer and Mercer, 1979) or variations in the diameter of existing ultrafine aerosols to which the $^{218}PoO_2^+$ ion can attach.

Clustering of both water vapor molecules (Raabe, 1968; Porstendorfer and Mercer, 1979; Hawrynski, 1984; Raes, 1985; Raes, et al., 1985) and of sulfuric acid (Chu, et al., 1987; Hopke and Ramamurthi,1988; Raes, et al., 1985) have been investigated. Raes (1985) estimated the lifetime of the charged Po ion to be of the order of seconds at typical indoor air concentrations of radon. He concluded that stable H_2O clusters will be rapidly formed around the charged Po ion before neutralization occurs under typical indoor conditions. Chu, et al. (1987) reported the formation of an ultrafine aerosol (0.5 to 3 nm) induced by radon radiolysis of a mixture of water vapor and SO_2 in air and subsequent clustering of sulfuric acid molecules around the Po-218 ion.

To date, there has been little consideration of the possible role of airborne organic compounds in ion-induced nucleation, although Keesee and Castleman (1989) have discussed the potential for forming molecular clusters from organic compounds in the troposphere. Specifically, these investigators considered reactions of nitrogen-containing organics with high proton affinities with tropospheric ion clusters. They did not evaluate the formation of clusters of indoor air organic compounds around charged ions formed by radon decay.

Indoor atmospheres contain many vapor-phase organic compounds which may interact with radon progeny to form ion clusters. Over 300 volatile organic compounds (VOC) have been detected in indoor air (Berglund, et al., 1986). The VOC are generally defined as organic compounds with vapor pressures greater than approximately 10^{-3} torr at 25 ^{o}C; this category includes compounds such as the C_4 to C_{16} alkanes, aromatic hydrocarbons such as benzene and toluene, chlorinated hydrocarbons, alcohols, aldehydes, ketones, esters, terpenes, ethers, and heterocyclics. Concentrations of individual VOC in indoor air are typically of the order of $\mu g/m^3$ but can range up to 100 times higher depending upon indoor VOC sources and ventilation rates. Concentrations of total VOC (TVOC = C_4 through about C_{16}) in indoor air range from a few milligrams per cubic meter to as much as 25 mg/m^3 (Lebret, et al., 1986; Molhave and Moller, 1979; DeBortoli, et al., 1985, 1986;Hodgson et al., 1989). By comparison, concentrations of SO_2 are typically less than 15 $\mu g/m^3$ in U.S. homes (Spengler and Sexton, 1983), unless there is a source such as a kerosene heater present. Semi-volatile organic compounds (SVOC), such as polycyclic aromatic hydrocarbons, are also found in indoor environments (Wilson, et al., 1985; Wilson and Chuang, 1987; Daisey et al., 1989; Offermann et al., 1990). These compounds have vapor pressures between about 10^{-4} and 10^{-8} torr and are found in both the vapor and particulate phases. Indoor concentrations of individual SVOC such as the polycyclic aromatic hydrocarbons are typically of the order of ng/m^3 (Wilson and Chuang, 1987; Daisey et al., 1989; Offermann et al., 1990); concentrations of total SVOC are not available but are probably of the order of tenths of milligrams per m^3 to a few mg/m^3.

The purpose of this study was to investigate the potential for cluster formation induced by the decay of radon in indoor air containing VOC and SVOC and to determine which VOC and SVOC were most likely to form clusters around the PoO_2^+ ion formed by radon decay. In addition, it was felt that a compilation of the indoor concentrations and properties of VOC and SVOC might be of value to the radon research community for planning future experiments and for more detailed calculations.

APPROACH

Although the formation of clusters of vapors and gases about ions is essentially an atomic/molecular-level process, the classical theory of charged liquid droplet formation (Thomson, 1888; Chan and Mohnen, 1980; Castleman, et al., 1978), which is based on the macroscopic properties of the clustering species, has often been used to describe this process (Castleman, 1982; Raes, 1985). In this theory, the Gibbs free energy of cluster formation around an ion is described by the Thomson equation:

$$\Delta G = -nkT \ln S + 4\pi r^2\sigma + (q^2/2)(1-1/\epsilon)\,(1/r - 1/r_0) \qquad (1)$$

where

ΔG = free energy required to form a cluster of radius, r, with an ion core of radius, r_0;
S = the saturation ratio = p/p_0;
p = ambient vapor pressure of an organic compound;
p_0 = saturation vapor pressure of an organic at temperature, T;
n = number of vapor molecules in the cluster;
k = the Boltzmann constant;
T = temperature in degrees K;
r = radius of the cluster;
σ = surface tension of the organic compound;
q = charge of the ion;
ϵ = dielectric constant of the organic compound;
r_0 = radius of the PoO_2^+ ion, 0.23 nm (Hammon, et al., 1975; Cotton and Wilkinson, 1988).

The variables n and r are related by:

$$4/3\ \pi r^3\rho = nM/N, \qquad (2)$$

where ρ is the bulk density of the clustering organic compound, M is its molecular weight and N is Avogadro's number.

The first term on the right of the equation accounts for the latent heat of condensation and is positive unless the condensing vapor is present in the atmosphere at supersaturation levels. The second term in the equation represents the free energy barrier due to the surface tension of the liquid droplet and is also positive. The last term of the equation contributes to the lowering of the cluster free energy due to the presence of the ion and is negative. Stable clusters will form only if there is a negative minimum in ΔG. For typical indoor or outdoor atmospheres, the value of p is always smaller than p_0. Thus, the values of the first two terms of equation (1) are positive for typical indoor or outdoor atmospheric conditions. The value of third term, however, is negative and, therefore, in the presence of ions, nucleation might occur at less than supersaturation with respect to VOC or SVOC. Despite the use of macroscopic

properties, Castleman (1982) has shown that the classical theory is a useful tool to quantitatively describe clustering of water molecules around various ions. For other species, the picture given by this theory is probably only qualitative.

In this investigation, the classical charged liquid drop nucleation theory was used as a first order approach to evaluate the potential for indoor air VOC and SVOC to form clusters around $^{218}PoO_2^+$. Since the equation that results from differentiating equation (1) and setting $\delta\Delta G/\delta n$ equal to zero cannot be easily solved in closed form, equation (1) was solved for a series of values of r and the minimum estimated to within about 0.1 Angstrom. The maximum reported indoor concentration was used to calculate ΔG for each VOC and SVOC for which physical properties were available or could be reasonably estimated. Calculations for water vapor (50% relative humidity) were also included for comparison to the VOC and SVOC.

Data on indoor concentrations of VOC and SVOC were taken largely from the compilation of McCann, et al. (1988) which covered much of the literature up to about 1984. These concentrations are consistent with indoor air measurements made at later dates (Shah and Heyerdahl, 1988). Additional data on indoor concentrations were compiled through a survey of the literature. The maximum and mean (or median) concentrations for 44 indoor air organic compounds are summarized in Table 1 and the sources of the measurements are given.

Indoor data for some compounds were quite sparse e.g., the polycyclic aromatic hydrocarbons (PAH). The maximum naphthalene value reported by Hawthorne, et al. (1984) was used for calculation since their data provide the largest single available data set, i.e., 40 homes. Higher indoor concentrations of naphthalene have since been reported. Offermann, et al. (1990) have reported a maximum indoor concentration of 3.3 $\mu g/m^3$. Brown, et al. (1990) have reported an exceptionally high indoor concentration of 970 $\mu g/m^3$ for a house in England which had been damp-proofed with a bituminous material containing coal tar. For the PAH, the median and maximum concentrations do not differ by much. This is probably due to the very limited set of measurements that have been made. As additional measurements are made for this compound class, it is likely that maxima greater than those listed in Table 1 will be reported.

Limited indoor concentration data were also available for 2-ethoxyethyl acetate; the maximum concentration reported by DeBortoli, et al. (1986), based on measurements in 6 houses, was used. For phenol, the data reported by Kalinic, et al. (1987) for 18 classrooms in Yugoslavia are reported in the table. These data are consistent with measurements reported by Wang (1975) for a college auditorium.

For nicotine, a common indoor contaminant from cigarette smoke, the measurements reported by McCarthy, et al. (1987) for 68 homes with smokers and 13 homes of non-smokers were combined with data reported by Mieser, et al. (1989) for other indoor environments to estimate a median concentration. The maximum concentration reported for these two data sets was 26.5 $\mu g/m^3$.

For the pesticides, chlordane and chlorpyrifos, medians were estimated from the data reported by Olds (1987). Olds (1987) reports results of measurements made in 4368 army base homes. It is reasonable to expect that similar results would be found for non-army homes.

Values of saturation vapor pressures, surface tension, dielectric constant, density and molecular weight were compiled from the literature and are summarized in Table 2. These values were estimated for some VOC and SVOC for which values of physical

constants were not available but for which data existed for similar compounds. The values of the saturation vapor pressures, usually at 25°C, were taken from Mackay and Shiu (1981) for the alkanes, the aromatic hydrocarbons (except styrene), naphthalene, 1-methylnaphthalene, biphenyl, the chlorinated hydrocarbons and chlorpyrifos. Equilibrium vapor pressures (20°C) for anthracene, phenanthrene, pyrene and fluoranthene, as solids, and for chlordane, a liquid, were from Foreman and Bidleman (1987). Other vapor pressures were obtained from the *CRC Handbook of Chemistry and Physics*, 70th Edition (1989), *Lange's Handbook of Chemistry* (1985) (cyclohexylamine), or *Hawley's Condensed Chemical Dictionary* (1987)(2-ethoxyethyl acetate). The vapor pressures for biphenyl, phenol and pentachlorophenol were extrapolated from temperatures at which these compounds are liquids to below their freezing points (i.e., supercooled liquids), and thus, are less certain than measured vapor pressures for solids. Vapor pressures for solids, however, are generally lower than those estimated for supercooled liquids (Mackay, *et al.*, 1982). Thus, the effect of this overestimate of the saturation vapor pressure on the calculated value of ΔG will be to make it more positive. The saturation vapor pressures for hexanal and nonanal were estimated from a plot of the logs of the vapor pressures of the C_3, C_4, C_7 and C_{11} aldehydes versus number of carbons.

Densities for all but one of the organic compounds were taken from the *CRC Handbook* (1989) or *Lange's Handbook of Chemistry* (1985) and are for 25°C or for 20°C. The density of chlordane was obtained from *Hawley's Condensed Chemical Dictionary* (1987). Most of the dielectric constants of the 44 compounds (generally for 25°C) in Table 2 were obtained from *Lange's Handbook of Chemistry* (1985) or the *CRC Handbook* (1989) or were estimated based on the dielectric constants of similar compounds. The dielectric constants of tetradecane and hexadecane were estimated to be about 2 based on the dielectric constants of the other alkanes. That of anthracene was estimated to be identical with that of phenanthrene; those of pyrene and fluoranthene were estimated to be slightly higher than the dielectric constant reported for phenanthrene. The dielectric constant for chlordane was estimated to be the same as that of chlorobenzene; the dielectric constant of chlorpyrifos, which has a pyridyl moiety in its structure, was taken as that of pyridine; that for cyclohexylamine was estimated from that of cyclohexanol and several amines. Since dielectric constants within series of related compounds typically vary by less than a $\pm$ 50%, these estimates are probably adequate for the purposes of the nucleation calculations. For example, the dielectric constants of the C_{10} through C_{12} alkanes vary from 1.99 to 2.015. Dielectric constants for the aromatic compounds range from 2.27 for benzene to only 2.57 for o-xylene.

Values of surface tension (at 25°C) were generally taken from the compilation of Jasper (1972). The surface tensions for naphthalene, tetrachloroethylene, p-dichlorobenzene, phenol, 2-ethoxyethyl acetate, pinene, and quinoline were taken from *Lange's Handbook of Chemistry* (1985). Surface tension data were not available for PAH other than naphthalene; thus, the surface tensions for these compounds were estimated to be the same as that of naphthalene. Similarly, the surface tension of pentachlorophenol was estimated to be identical with that of phenol; that of limonene was estimated to be the same as that of α-pinene, which is an isomer; that of chlordane was estimated to be the same as that of chlorobenzene; that of chlorpyrifos was estimated to be the same as that of pyridine. The surface tension of cyclohexylamine was estimated from those of ethanol and diethylamine. The surface tensions of hexanal and nonanal were estimated by interpolation between surface tensions of related aldehydes. The measured surface tensions for the 44 compounds listed in Table 2 range from 23 to 40 dynes/cm, a range of less than a factor of two. Surface tensions for related compounds (See Table 2) vary even less. Thus, errors in the estimated values are expected to be relatively small.

There were many additional VOC and SVOC for which indoor measurements were available but physical properties (saturation vapor pressure, surface tension, or dielectric constant) were not available and could not be reliably estimated. These compounds could not be included in this assessment.

RESULTS

Table 3 summarizes the values calculated for the minimum ΔG (ΔG_m) and the estimated average number of molecules per cluster. The molecular weights, saturation vapor pressures and maximum indoor concentration used in the calculations are also presented here for easy reference. The minimum values of ΔG were all negative with the exceptions of those of benzene, naphthalene, 1-methylnaphthalene, and tetrachloroethylene. The compounds with the most negative ΔG_m values ($<-4 \times 10^{-13}$ ergs) were largely found in the oxidized hydrocarbon class (e.g., n-butanol, i-butanol, phenol, hexanal, nonanal, benzaldehyde, the ketones and the acetates) and the SVOC class (pentachlorophenol, nicotine, chlordane, chlorpyrifos). Water vapor, in comparison, had a value of -4.3×10^{-13} ergs per cluster. The Thomson equation estimates a radius of 0.36 nm and 6.5 water molecules per cluster, which is consistent with the diffusion coefficients which have been measured (Raabe, 1968; Raes, 1985). For the VOC and SVOC, the radii (diameters) at the minimum ΔG_m ranged from 0.32 nm to 0.49 nm (0.6 nm to 1.0 nm). The charged liquid drop theory thus suggests clusters with diameters that fall within the measured range of diameters for the "unattached" fraction, i.e., 0.5 to 3 nm.

Table 4 presents the the values of each of the three terms that contribute to the total ΔG_m for those VOC and SVOC with the most negative ΔG_m values. ΔG_1 is the latent heat of condensation, which, with the exception of chlordane, is always positive for the VOC and SVOC in indoor air. ΔG_2 is the surface energy term which is also positive and ΔG_3 is the change in the electric field energy, which is always negative. For the VOC and SVOC with the most negative values of ΔG_m, the first two terms summed are all less than 12×10^{-13} ergs while the third term, which is negative, ranges from -14×10^{-13} to -23.5×10^{-13} ergs. The oxidized hydrocarbons tend to have relatively high values for their dielectric constants which contribute to larger negative values of ΔG_3. For the SVOC, the ratios of indoor air concentrations to saturation vapor pressures are generally larger than for the VOC. Thus, the first term in equation (1) is somewhat less positive for the SVOC than the VOC.

For some of the compounds in Table 4, the input values of surface tension and dielectric constant had to be estimated: hexanal, nonanal, pentachlorophenol, nicotine (ϵ only), chlordane and chlorpyrifos. The effects of errors in the estimated values of the dielectric constant on ΔG_m will be relatively small because of the inverse relationship between ΔG_3 and ϵ. Within a class of compounds, the error in the estimated values of ϵ is probably not larger than 50%, at most. This error would have the greatest impact for the smallest value of estimated ϵ, that of nonanal (5.5). If this value were 50% lower (2.75), the value of the $(1-1/\epsilon)$ term would only be reduced to 0.636 (-22%) relative to the value calculated at 5.5 (0.818). If ϵ were actually 50% higher, this term would be increased only to 0.879. Thus, the uncertainties in the estimated values of ϵ will not have a very substantial impact on the values of ΔG_3. Errors in the estimated values of σ will have a more direct effect on ΔG_2 and ΔG_m. The values of σ for all of the compounds in the tables vary by less than a factor of 2 (23 to 40). Within a class of compounds, the errors in the estimates of σ are probably no more than 50%. For example, the measured value of σ for butanal is 24.36 while that for heptanal is 26.34. A 50% increase (decrease) in the value of σ would increase (decrease) the value

of ΔG_2 by 50%. A 50% increase in ΔG_2 for the compounds for which σ was estimated in Table 4 would not be large enough to make ΔG_m positive.

The values of ΔG_m for ion-induced nucleation were all calculated on the basis of the maximum indoor concentration found in the literature. Ratios of maximum to median or mean concentration range from as low as about 2 to a high of 325 for chlorpyrifos, but are typically less than 100. Table 5 compares the values of ΔG_m and its components for the maximum and mean concentrations for several of the VOC and SVOC for which this ratio is high. In none of these cases does the value of ΔG_m become positive; the overall change is typically only about 2.5 x 10^{-13} ergs per cluster. The value of ΔG_1, which incorporates the term ln p/p_0, is most affected by a change in concentration; however, even for a reduction in concentration from 87.7 $\mu g/m^3$ to 0.27 $\mu g/m^3$ (325-fold), the value of ΔG_1 is increased only by a factor of four (0.64 x 10^{-13} to 2.53 x 10^{-13}) because of the logarithmic relationship. The values of ΔG_2 and ΔG_3 also change slightly with concentration due to small changes in the minimum radius. For example, for chlorpyrifos, the radius at the minimum for ΔG is 0.47 nm for the maximum concentration and 0.43 nm at the mean concentration. Thus, even if the median rather than the maximum concentrations were used for these calculations, the values of ΔG_m and r_m would not change substantially.

DISCUSSION

The classical charged liquid droplet theory (Thomson equation) predicts that stable clusters of $^{218}PoO_2^+$ with VOC and SVOC will form under typical indoor air conditions. This theory also predicts that the clusters will consist of one molecule of organic with each ion. The values of ΔG calculated from this theory rise steeply and become positive for values of r greater than about 0.5 nm ($n > 1$) in all of the cases examined here. While the quantitative predictions of the values of ΔG are not likely to be correct, it is of interest to ask whether the qualitative prediction, that stable clusters of VOC and SVOC with $^{218}PoO_2^+$ will be formed under typical indoor environment conditions, is likely to be correct.

The case for water clusters can provide some useful insights. The work of Castleman (1982) suggests that the classical theory gives reasonable predictions for the clustering of water molecules about alkali cations for clusters greater than about 6 or 7 molecules. This theory predicts formation of a stable cluster for six water molecules for the $^{218}PoO_2^+$ ion at 50% relative humidity. This prediction is consistent with the diffusion coefficients which have been reported for the polonium ion in the presence of water vapor (Raabe, 1968; Raes, 1985).

The classical theory is based on the bulk properties of liquids at a macroscopic level and does not take into account effects at the molecular level. This is the level at which clustering initially occurs, i.e., one or two molecules clustered about an ion. Chan and Mohnen (1980) developed an empirical-analytical semi-molecular theory of the nucleation of water about an ion to try to correct for molecular level effects. In this model, the Thomson equation is used to calculate ΔG for clusters of water molecules about an ion, but the first shell of hydration, assumed to consist of the first six water molecules, is treated separately. Since the classical theory neglects the translation and rotation of the microdroplet and the heat of hydration, a correction is made to the first term of the Thomson equation by incorporating these terms for the clusters consisting of 1 to 6 molecules about the ion. The Chan and Mohnen model also corrects for the change in surface tension as the droplet becomes smaller, for the change in the electric field due to the condensation of a dielectric film about the ion, and for surface dipole-dipole interactions. The overall effect of these corrections for

water molecules clustered about a Cs^+ ion is to approximately double the negative value of ΔG which is calculated using the classical theory, i.e., ΔG becomes more negative by a factor of about 2 over the the curve for ΔG versus n. These results suggest that the values of ΔG calculated for clusters of VOC and SVOC about the $^{218}PoO_2^+$ using the Thomson equation may underestimate the stability of the clusters.

Castleman and coworkers (1978) have compared the predictions of the classical droplet theory and of some experimental measurements of clusters of water and ammonia about some cations. For the Bi^+ ion, which most closely resembles polonium in terms of its chemistry, the Thomson equation predicted less negative values for the enthalpy of cluster formation with NH_3 than were observed experimentally for values of n from 1 to about 6. No experimental data were available for the Bi^+ ion with water. The experimental data for clusters of NH_3 with the Bi^+ ion suggested also that the entropy of formation of these microclusters was more negative than predicted by theory, that is, the clusters have a more ordered structure than accounted for in the classical liquid droplet theory. Overall, the experimental results for clusters of a few molecules of NH_3 with the Bi^+ ion indicate that ΔG for cluster formation is more negative (more stable cluster) than predicted by the classical theory.

Thus, the limited experimental data which are available and the results of more detailed theoretical calculations for water (or NH_3) molecules clustered about cations other than $^{218}PoO_2^+$ suggest that the qualitative prediction of formation of stable clusters of VOC and SVOC with this ion, based on the classical liquid droplet theory, is probably correct, but that the magnitude of the ΔG_m calculated may not be sufficiently negative.

The classical charged liquid droplet theory predicts that the oxidized hydrocarbons and semi-volatile organic compounds in indoor air are the most likely to form clusters with the $^{218}PoO_2^+$ ion. The macroscopic properties of the VOC and SVOC reflect the molecular level properties of these compounds relative to the non-polar hydrocarbons. The compounds in these classes all have permanent dipole moments and/or relatively low equilibrium vapor pressures. Consequently, they are more likely to interact with an ion than are the non-polar hydrocarbon compounds. Thus, the predictions of the simple liquid droplet theory with respect to the compounds most likely to form clusters are quite reasonable.

The Thomson equation also predicts the formation of clusters consisting of a single molecule of organic with each ion, with cluster diameters of the order of 0.6 to 1.0 nm. Such a simple dimer <u>does</u> meet the definition of a cluster as an aggregate of as few as two species as defined by Castleman and Keesee (1988). Although the predicted cluster diameters are consistent with measured diameters for the "unattached" fraction in homes, there is not sufficient experimental information to judge the validity of this prediction. More detailed calculations are unlikely to adequately address this question given the current state of development of theory for such clusters. Thus, experimental investigations of cluster formation are recommended to confirm the qualitative predictions of the Thomson equation and to make quantitative determinations of cluster size. The VOC and SVOC identified in this investigation as most likely to form clusters could be used in such experiments.

Existing theoretical treatments of cluster formation examine the clustering of a single species (e.g., water, acetone) around a given ion. In this study, only 44 individual compounds out of the hundreds of VOC and SVOC which have been found in indoor air were examined. The "real world" of indoor air is considerably more complex and contains mixtures of water vapor, VOC and SVOC. The more polar organic compounds are likely to interact with each other and with water molecules

clustered around ions. Given the present state of our knowledge, such complex interactions are also best investigated experimentally.

The prediction of cluster formation of the $^{218}PoO_2^+$ ion with VOC and SVOC found in indoor air has implications for experimental determinations of the diffusion coefficient of this ion. Such experiments have often been conducted with unpurified outdoor or indoor air. Since ambient air always has VOC and SVOC present, it should be purified to remove these compounds in any determinations of diffusion coefficients. Furthermore, sources of VOC and SVOC contaminants in the experimental apparatus should be considered. For example Tygon tubing, which is widely used in laboratories, is a source of phthalate esters (SVOC) which have some potential for cluster formation.

SUMMARY AND CONCLUSIONS

The purpose of this study was to investigate the potential for cluster formation induced by the decay of radon in indoor air containing vapor phase organic compounds and to determine which were most likely to form clusters around the PoO_2^+ ion formed by radon decay. The classical charged liquid droplet theory (Thomson equation) was used to estimate the Gibbs free energy of ion-induced nucleation and to provide an indication of the indoor organic compounds most likely to undergo ion-induced nucleation.

The results indicate that there is a potential for the formation of ion-induced clusters of organic compounds around the $^{218}PoO_2^+$ ion. The compounds with the greatest potential for cluster formation are the oxidized hydrocarbons (e.g., n-butanol, phenol, hexanal, nonanal, benzaldehyde, the ketones and the acetates) and the semi-volatile organic compounds (pentachlorophenol, nicotine, chlordane, chlorpyrifos). The theory estimates an average of one VOC molecule per ion and cluster diameters of 0.6 to 1.0 nm. Although these estimated diameters are consistent with measured diameters for the "unattached" fraction in homes, the state of experimental and theoretical knowledge with respect to cluster formation with VOC and SVOC is not adequate to judge the validity of this prediction. Given the current state of theoretical knowledge, experimental determinations are recommended .

ACKNOWLEDGMENTS

This research was supported the Director, Office of Energy Research, Office of Health and Environmental Research, Human Health and Assessments Division, U.S. Department of Energy, under Contract No. DE-AC03-76SF00098. Helpful discussions of this work and careful reviews of the manuscript by P. Hopke, W.W. Nazaroff and C. Weschler are gratefully acknowledged.

REFERENCES

Berglund, B., U. Berglund, and T. Lindvall. (1986). Assessment of discomfort and irritation from the indoor air. *Proceedings IAQ '86: Managing Indoor Air for Health and Energy Conservation*, ASHRAE, Atlanta, GA, pp. 138-149.

Brown, V.M., A.H. Cockram, D.R. Crump and D. Gardiner. (1990). Investigations of the volatile organic compound content of indoor air in homes with an odorous damp proof membrane. In: *Indoor Air '87, Proceedings of the 4th International Conference on Indoor Air Quality and Climate*, Seifert, B., et al., Eds., Berlin (West), 17-21 August, 1987, Institute for Water, Soil and Air Hygiene, Berlin , Vol. 3, pp. 575-580.

Castleman, Jr., A.W. (1982). Experimental studies of ion clustering: relationship to aerosol formation processes and some atmospheric implications. *J. Aerosol Sci.*, **13**: 73-

Castleman, Jr., A.W., P.M. Holland, and R.G. Keesee. (1978). The properties of ion clusters and their relationship to heteromolecular nucleation. *J. Chem. Phys.*, **68**: 1760-1767.

Castleman, Jr., A.W. and R.G. Keesee. (1988). Gas-phase clusters: Spanning the states of matter. *Science*, **241**: 36-42.

Chan, L.-Y. and V.A. Mohnen. (1980). Ion nucleation theory. *J. Atmos. Sci.*, **37**: 2323-2331.

Chu, K.-D., P.K. Hopke, E.O. Knutson, K.W. Wu and R.F. Holub. (1987). Induction of an ultrafine aerosol by radon radiolysis. In: *Radon and its Decay Products, Occurrence, Properties and Health Effects*, P.K. Hopke, Ed., ACS Symposium Series No. 331, American Chemical Society, Washington, D.C., pp. 365-376.

Cotton, F.A. and G. Wilkinson. (1988). *Advanced Inorganic Chemistry*, Fifth Edition. John Wiley and Sons, Inc., New York, 1455 pp.

CRC Handbook of Chemistry and Physics, 70th Edition. (1989). R.C. Weast, D.R. Lide, M.J. Astle and W.H. Beyer, Eds. CRC Press, Inc., Boca Raton, FL

Daisey, J.M., J.D. Spengler, and P. Kaarakka. (1989). A comparison of the organic chemical composition of indoor aerosols during woodburning and non-woodburning periods. *Environ. Int.*, **15**: 435-442.

De Bortoli, M., H. Knoppel, E. Pecchio, A. Peil, L. Rogora, H. Schauenburd, H. Schlitt and H. Vissers. (1985). Measurements of indoor air quality and comparison with ambient air. EUR 1656 EN, Directorate-General for Science, Research and Development, Joint Research Center.

De Bortoli, M., H. Knoppel, E. Pecchio, A. Peil, L. Rogora, H. Schauenburd, H. Schlitt and H. Vissers. (1986). Concentrations of selected organic pollutants in indoor and outdoor air in northern Italy. *Environ. Int.*, **12**: 343-350.

Edgerton, S.A., D.V. Kenny and D.W. Joseph. (1989). Determination of amines in indoor air from steam humidification. *Environ. Sci. Technol.*, **23**: 484-488.

Foreman, W.T. and T.F. Bidleman. (1987). An experimental system for investigating vapor-particle partitioning of trace organic pollutants. *Environ. Sci. Technol.*, **21**: 869-

875.

Hammon, H.G., K. Ernat, J.R. Gaskill, J.C. Newton, and C.J. Morris. (1975). Development and evaluation of radon sealants for uranium mines. Lawrence Livermore Laboratory, Livermore, CA,94550, Report No. UCRL-51818, TID-4500, UC-41, May 29, 1975.

Hartwell, T.D., R.L. Perritt, H.S. Zelon, R.W. Whitmore and E.D. Pellizzari. (1984). Comparisons of indoor and outdoor levels for air volatiles in New Jersey. *Proceedings of the 3rd International Conference on Indoor Air Quality and Climate*, Stockholm, Sweden, pp. 57-61.

Hawley's Condensed Chemical Dictionary. 11th Edition. (1987). Revised by N.I. Sax and R.J. Lewis, Sr. Van Nostrand - Reinhold Company, New York, NY.

Hawrynski, M.J. (1984). The theory of clusters. In: *Occupational Radiation Safety in Mining: The Proceedings of the International Conference*, Vol.2, pp. 551-560.

Hawthorne, A.R., R.B. Gammage, C.S. Dudney, T.G. Matthews, and D.J. Erdman. (1984). Formaldehyde levels in forty east Tennessee homes, In: *Proceedings of the 3rd International Conference on Indoor Air Quality and Climate*, Stockholm, Sweden, Vol. 3, pp.17-22.

Hodgson, A.T., J.M. Daisey, and R. Grot. (1989). Sources and source strengths of volatile organic compounds in a new office building. *J. Air Waste Manag. Assoc.* (In Press). LBL-26950.

Hopke, P.K. (1987). The indoor radon problem explained for the layman. In: *Radon and its Decay Products, Occurrence, Properties and Health Effects*, P.K. Hopke, Ed., ACS Symposium Series No. 331, American Chemical Society, Washington, D.C., pp. 572-586.

Hopke, P.K. and Ramamurthi. (1988). Production of ultrafine particles by radon radiolysis. *J. Aerosol Sci.*, **19**: 1323-1325.

Jasper, J.J. (1972). The surface tension of pure liquid compounds. *J. Phys. Chem. Ref. Data*, **1**: 841-945.

Kalinic, N., K. Sega, J. Hrsak, and A. Sisovic. (1987). Phenol levels in indoor air. In: *Indoor Air '87, Proceedings of the 4th International Conference on Indoor Air Quality and Climate*, Seifert, B., et al., Eds., Berlin (West), 17-21 August, 1987, Institute for Water, Soil and Air Hygiene, Berlin , Vol. 1, pp. 235-238.

Keesee, R.G. and A.W. Castleman, Jr. (1989). The properties of organic compounds in molecular clusters. *Aerosol Sci. Technol.*, **10**: 352-357.

Krause, C., W. Mailahn, R. Nagel, C. Schulz, B. Seifert and D. Ullrich. (1987). Occurrence of volatile organic compounds in the air of 500 homes in the Federal Republic of Germany. In: *Indoor Air '87, Proceedings of the 4th International Conference on Indoor Air Quality and Climate*, Seifert, B., et al., Eds., Berlin (West), 17-21 August, 1987, Institute for Water, Soil and Air Hygiene, Berlin, Vol. 1, pp. 102-106.

Lange's Handbook of Chemistry. (1985). 13th edition. John A. Dean, Ed. McGraw-Hill Book Co., New York, NY.

Lebret, E., H.J. van de Wiel, H.P. Bos, D. Noij and J.S.M. Boleij. (1986). Volatile organic compounds in Dutch homes. *Environ. Int.*, **12**: 323-332.

Mackay, D., A. Bobra, D. W. Chan, and W. Y. Shiu. (1982). Vapor pressure correlations for low volatility environmental chemicals. *Environ. Sci. technol.*, **16**: 645-649.

Mackay, D. and W.Y. Shiu. (1981). Critical review of Henry's Law constants for chemicals of environmental interest. *J. Phys. Chem. Ref. Data*, **10**: 1175-1199.

McCann, J., L. Horn, J. Girman and A.V. Nero. (1988). Potential risks from exposure to organic compounds in indoor air. Lawrence Berkeley Laboratory Report LBL-22473, Berkeley, CA.

McCarthy, J., J. Spengler, B.-H. Chang, D. Coultas and J. Samet. (1987). A personal monitoring study to assess exposure to environmental tobacco smoke. In: *Indoor Air '87, Proceedings of the 4th International Conference on Indoor Air Quality and Climate*, Seifert, B., et al., Eds., Berlin (West), 17-21 August, 1987, Institute for Water, Soil and Air Hygiene, Berlin, Vol. 2, pp. 142-146.

Miesner, E.A., S.N. Rudnick, F.-C. Hu, J.D. Spengler, H. Ozkaynak, L. Preller and W. Nelson. (1989). Particulate and nicotine sampling in public facilities and offices. *J. Air Waste Manag. Assoc.*, **39**: 1577-1582.

Molhave, L. and J. Moller. (1979) The atmospheric environment in modern Danish dwellings - measurements in 39 flats. In: *Indoor Air*, P.O. Fanger and O. Valbjorn, Eds., pp. 171-186, SBE, Horsholm, Denmark.

Monteith, D.K., T.H. Stock and J.E. Seifert. (1984). Sources and characterization of organic air contaminants in manufactured housing. In: *Proceedings of the 3rd International Conference on Indoor Air Quality and Climate*, Stockholm, Sweden, Vol. 4, pp. 285-290.

Offermann, F.J., S.A. Loiselle, J.M. Daisey, A.T. Hodgson, and L.A. Gundel. (1990). *Sampling, Analysis, and Data Validation of Indoor Concentrations of Polycyclic Aromatic Hydrocarbons*. Final Report, Contract No. A732-106, prepared for the California Air Resources Board, March, 1990.

Olds, K.L. (1987). Indoor airborne concentrations of termiticides in Department of the Army housing. In: *Indoor Air '87, Proceedings of the 4th International Conference on Indoor Air Quality and Climate*, Seifert, B., et al., Eds., Berlin (West), 17-21 August, 1987, Institute for Water, Soil and Air Hygiene, Berlin, Vol. 1, pp. 205-209.

Porstendorfer, J.A. and T.T. Mercer. (1979). Influence of electric charge and humidity upon the diffusion coefficient of radon decay products. *Health Physics*, **37**: 191-199.

Raabe, O. (1968). Measurement of the diffusion coefficient of radium A. *Nature (London)*, **217**: 1143 -1145.

Raes, F. (1985) Description of the properties of unattached ^{218}Po and ^{212}Pb particles by means of the classical theory of cluster formation. *Health Phys.*, **49**: 1177-1187.

Raes, F., A. Janssens and H. Vanmarcke. (1985). A closer look at the behaviour of radioactive decay products in air. *Sci. Total Environ.*, **45**: 205-218.

Reineking, A., K.H. Becker and J. Porstendorfer. (1988). Measurements of activity size distributions of the short-lived radon daughters in the indoor and outdoor environment. *Radiat. Prot. Dosim.*, **24**: 245-250.

Sauer, J.M., P.J. Walcheski, D.D. Nicholas, and L.R. Gjovik. (1982). The concentration of airborne pentachlorophenol within treated wood structures. *Proceedings of the American Wood-Preservers Association Annual Meeting*, New Orleans, LA. Vol. 78, pp.169-173.

Shah, J.J., and E.K. Heyerdahl. (1988). *National Ambient Volatile Organic Compounds (VOCs) Data Base Update.* EPA 600/3-88/010(a). U. S. Environmental Protection Agency, Research Triangle Park, NC. NTIS no. PB 88-195 631/AS, PB88-189 022/AS.

Spengler, J.D. and K. Sexton. (1983). Indoor air pollution: A public health perspective. *Science*, **221**: 9-16.

Strong, J.C. (1988). The size of attached and unattached radon daughters in room air. *J. Aerosol Sci.*, **19**: 1327-1330.

Thomson, J.J. (1888) *Application of Dynamics to Physics and Chemistry*, 1st ed., Cambridge University Press.

Tu, K.W., E.O. Knutson and A.C. George. (1991). Indoor radon progeny aerosol size measurements in urban, suburban and rural regions. *Aerosol Sci. Technol.*, **15**:170-178.

Wallace, L., et al. (1982). Monitoring individual exposure. Measurements of volatile organic compounds in breathing zone air, drinking water, and exhaled breath. *Environ. Intern.*, **8**: 269-282.

Wang, T.C. (1975). A study of bioeffluents in a college classroom. *ASHRAE Transactions*, **81 (part 1)**: 32-44.

Wilson, N.K., R.G. Lewis, C.C. Chuang, B.A. Petersen and G.A. Mack. (1985). Analytical and sampling methodology for characterization of polynuclear aromatic compounds in indoor air. *Proceeding of the 78th Annual Meeting of the Air Pollution Control Association*, Detroit, Michigan, June 16-21, 1985.

Wilson, N.K. and J.C. Chaung. (1987) Indoor air levels of polynuclear aromatic hydrocarbons and related compounds in eight-home pilot study. In: *Proceedings of the 11th International Symposium on Polynuclear Aromatic Hydrocarbons*, Gaithersburg, MD, September, 1987.

Table 1. Maximum and Mean (or Median) Measured Indoor Concentrations of Some Volatile and Semi-Volatile Organic Compounds

Compound	n[a.]	Concentrations, ug/m^3		Reference
		Maximum	Mean or Median	
Alkanes				
n-Decane	46	2770	42	Molhave, et al., 1979
u-Undecane	46	2360	670	Molhave, et al., 1979
n-Dodecane	40	675	9	Hawthorne, et al., 1984
n-Tetradecane	40	74	5.4	Hawthorne, et al., 1984
n-Hexadecane	40	21	3.8	Hawthorne, et al., 1984
Aromatic Hydrocarbons				
Benzene	17	387	4.6	Wallace, et al., 1982
Toluene	134	700	55	Lebret, et al., 1984
Styrene	85	54	1.8	Hartwell, et al., 1984
Ethylbenzene	85	320	6.1	Hartwell, et al., 1984
p-Xylene	2	294	50	Pellizzari, et al., 1984
m-Xylene	46	910	145	Molhave, et al., 1979
o-Xylene	15	132	17	De Bortoli, et al., 1985
1,2,4-Trimethylbenzene	46	1140	170	Molhave, et al., 1979
n-Butylbenzene	134	40	0.9	Lebret, et al., 1987
Polycyclic Aromatic Hydrocarbons				
Napthalene	40	0.675	0.013	Hawthorne, et al., 1984
1-Methylnaphthalene	134	0.0022	<0.0003	Lebret, et al., 1984
Biphenyl	5	0.572	0.405	Offermann, et al., 1984
Anthracene	9	0.046	0.006	Wilson, et al, 1985; 1987; Offermann, et al., 1990
Phenanthrene	9	0.210	0.066	Wilson, et al, 1985; 1987; Offermann, et al., 1990
Pyrene	9	0.036	0.007	Wilson, et al, 1985; 1987; Offermann, et al., 1990
Fluoranthene	9	0.037	0.008	Wilson, et al, 1985; 1987; Offermann, et al., 1990

a. n = number of buildings

Table 1. Maximun and Mean (or Median) Measured Indoor Concentrations of Some Volatile and Semi-Volatile Organic Compounds

Compound	n[a.]	Concentrations, ug/m^3		Reference
		Maximum	Mean or Median	
Chlorinated Hydrocarbons				
1,1,1-Trichloroethane	85	880	16	Hartwell, et al., 1984
Tetrachloroethylene	85	250	5.6	Hartwell, et al., 1984
p-Dichlorobenzene	85	915	2.8	Hartwell, et al., 1984
Oxygenates				
n-Butanol	44	80	16	Monteith, et al., 1984
i-Butanol	230	20	2	Krause, et al., 1989
Phenol	18	77.3	5	Kalinic, et al., 1987
Pentachlorophenol	104	25	5	Krause, et al., 1989
Butanal	15	34	<1	De Bortoli, et al., 1985
Hexanal	15	58	5	De Bortoli, et al., 1985
Nonanal	15	82	6	De Bortoli, et al., 1985
Benzaldehyde	40	124	19	Hawthorne, et al., 1984
Acetone	15	157	23	De Bortoli, et al., 1985
2-Butanone	15	38	4	De Bortoli, et al., 1985
Ethylacetate	230	204	6.8	De Bortoli, et al., 1985
2-Ethyoxyethylacetate	6	130	11	De Bortoli, et al., 1985
Other				
α-Pinene	46	830	315	Molhave, et al., 1979
β-Pinene	15	104	9	De Bortoli, et al., 1985
Limonene	15	480	57	De Bortoli, et al., 1985
Nicotine	100	26.5	1.9	McCarthy, et al, 1987; Mieser, et al., 1989
Quinoline	4	1.10	~0.022	Wilson, et al, 1985; Wilson and Chuer, et al., 1985
Chlordane	4368	130	1.82	Olds, 1987
Chlorpyrifos	4368	87.7	0.27	Olds, 1987
Cyclohexylamine	1	14.4	--	Edgerton, et al., 1989

[a.] n = number of buildings

Table 2. Properties of Some Indoor Organic Compounds

Compound	M.W.	SAT V.P.	Surf. Tens.	Diel. Const.	Density
	g.	mm Hg	dynes/cm		g/cm^3
n-Decane	148.28	1.32	23.37	1.99	.730
u-Undecane	156.32	.391	24.21	2.005	.741
Dodecane	170.33	.118	24.91	2.015	.766
Tetradecane	190.38	.00956	26.13	2.0[a.]	.765
Hexadecane	226.44	.0006757	27.05	2.0[a.]	.773
Benzene	78.11	95.56	28.22	2.274	.879
Toluene	92.13	28.1	27.93	2.379	.867
Styrene	104.14	7.18	31.6	2.426	.907
Ethylbenzene	106.17	9.88	28.75	2.412	.867
p-Xylene	106.17	8.74	28.01	2.27	.861
m-Xylene	106.17	8.28	28.47	2.374	.864
o-Xylene	106.17	6.54	29.76	2.568	.88
1,2,4-Trimethylbenzene	120.2	2.03	29.2	2.279	.865
n-Butylbenzene	134.2	1.026	28.7	2.36	.860
Napthalene	128.19	.082	40.07	2.52	1.145
1-Methylnaphthalene	142.2	.0665	40.0[a.]	2.71	1.025
Biphenyl	154.21	.00978	32.3	2.62[b.]	.866
Anthracene	178.23	.0000043	40.0[a.]	2.8[a.]	1.250
Phenanthrene	178.23	.000093	40.0[a.]	2.8	.980
Pyrene	202.26	.0000032	40.0[a.]	2.9[a.]	1.271
Fluoranthene	202.26	.0000051	40.0[a.]	2.9[a.]	1.252
1,1,1-Trichloroethane	133.4	124.4	34.02	3.42	1.456
Tetrachloroethylene	165.83	18.7	31.7	2.3	1.623
p-Dichlorobenzene	147.01	.679	32.46	3.41	1.458

[a.] Estimated, see text

[b.] Extrapolated from the liquid phase to below the freezing point

[c.] Interpolated from data on related aldehydes

Table 2. Properties of Some Indoor Organic Compounds

Compound	M.W.	SAT V.P.	Surf. Tens.	Diel. Const.	Density
	g.	mm Hg	dynes/cm		g/cm^3
n-Butanol	74.12	7.0	24.95	17.1	.810
i-Butanol	74.12	12.37	22.54	17.7	.808
Phenol	94.11	0.3[b.]	40.4	10.78	1.072
Pentachlorophenol	266.35	.00011[b.]	40.0[a.]	10.78	1.978
Butanal	72.1	111.0	24.36	13.4	.817
Hexanal	100.16	12.0[c.]	25.9[c.]	9.6[c.]	.814
Heptanal	114.18	.78	26.34	8.1	.850
Nonanal	142.24	.58[c.]	28.0[c.]	5.5[c.]	.826
Benzaldehyde	106.12	.93	38.0	17.8	1.050
Acetone	58.08	230.8	24.02	20.7	.792
2-Butanone	72.1	100.0	23.96	18.5	.805
Ethylacetate	88.1	94.7	23.39	6.02	.901
2-Ethyoxyethylacetate	132.16	2.0	31.8	7.6	.974
α-Pinene	136.24	4.41	26.0	2.64	.858
β-Pinene	136.24	2.95	25.92	2.76	.865
Limonene	136.24	2.1	26.0	2.3	.841
Nicotine	162.23	.004	38.29	10.0	1.009
Quinoline	129.15	0.1	39.59	9.0	1.093
Chlordane	409.8	.0000036	33.0[a.]	5.6[a.]	1.61
Chlorpyrifos	350.58	.0000187	38.0[a.]	12.3[a.]	1.5
Cyclohexylamine	99.18	8.7	31.22	4.73	.86
Water	18.02	23.756	71.97	78.54	.997

[a.] Estimated, see text

[b.] Extrapolated from the liquid phase to below the freezing point

[c.] Interpolated from data on related aldehydes

Table 3. Ion-Induced Nucleation of VOC in Indoor Air via Radon Decay

COMPOUND	M.W. (g)	SAT.V.P. x 10^{-6} (ppb)	MAX. CONC. (ppb)	ΔG_m [a.] x 10^{13} (ergs)	Avg. No. of Molecules in Cluster
OTHER CLASSES					
1,1,1-Trichloroethane	133.4	163.7	161	-0.5	0.9
Tetrachloroethylene	165.83	24.6	36.9	0.6	0.7
p-Dichlorobenzene	147.01	0.893	152	-3.3	1.1
n-Butanol	74.12	9.21	16.8	-6.2	1.3
i-Butanol	74.12	16.28	6.6	-5.6	1.3
Phenol	94.11	0.395	20.1	-4.4	1.3
Pentachlorophenol	266.34	0.000145	2.3	-9.3	1.4
Butanal	72.1	146	11.5	-0.4	1.1
Hexanal	100.16	15.8	14.1	-7.6	1.0
Nonanal	142.24	0.757	14.1	-7.6	0.9
Benzaldehyde	106.12	1.22	28.6	-5.8	1.3
Acetone	58.08	303.7	66.09	-3.4	1.4
2-Butanone	72.1	131.6	12.9	-4.6	1.2
Ethylacetate	88.1	124.6	56.6	-4.1	1.1
2-Ethoxyethylacetate	132.16	2.63	24	-6.6	1.1
a-Pinene	136.24	5.80	149	-3.8	0.8
b-Pinene	136.24	3.88	18.7	-3.6	0.8
Limonene	136.24	2.76	86.1	-2.9	0.7
Nicotine	162.23	0.005	4.0	-8.5	1.1
Quinoline	129.15	0.13	0.208	-4.2	0.9
Chlordane	409.8	0.00000467	7.76	-12.1	1.2
Chlorpyrifos	350.58	0.0000246	6.11	-12.3	1.0
Cyclohexylamine	99.18	11.45	3.5	-3.0	0.9

a. ΔG_m = Gibbs free energy at minimum

Table 3. Ion-Induced Nucleation of VOC in Indoor Air via Radon Decay

COMPOUND	M.W. (g)	SAT.V.P. x 10^{-6} (ppb)	MAX. CONC. (ppb)	ΔG_m [a.] x 10^{13} (ergs)	Avg. No. of Molecules in Cluster
ALKANES					
n-Decane	148.28	1.74	457	-3.3	0.7
Undecane	156.32	0.51	369	-3.6	0.7
Dodecane	170.33	0.16	98.9	-3.6	0.6
Tetradecane	190.38	0.0126	9.5	-3.5	0.6
Hexadecane	226.44	0.00089	2.27	-3.9	0.6
AROMATIC HYDROCARBONS					
Benzene	78.11	126	121	1.0	0.8
Toluene	92.13	37	186	-0.7	0.8
Styrene	104.14	9.45	12.7	-0.2	0.7
Ethylbenzene	106.17	13.0	81.8	-1.3	0.8
p-Xylene	106.17	11.5	67.7	-1.0	0.7
m-Xylene	106.17	10.9	209.6	-1.6	0.8
o-Xylene	106.17	8.60	30.4	-1.3	0.8
1,2,4-Trimethylbenzene	120.2	2.67	231.9	-2.0	1.2
n-Butylbenzene	134.2	1.35	7.3	-2.0	0.7
PAH[b.]					
Napthalene	128.19	0.108	0.13	0.8	0.7
1-Methylnapthalene	142.2	0.0875	0.002	0.4	0.6
Biphenyl	154.21	0.0129	0.091	-2.7	0.7
Anthracene	178.23	0.0000057	0.0063	-2.8	0.8
Phenanthrene	178.23	0.000122	0.0288	-2.9	0.6
Pyrene	202.26	0.0000042	0.0044	-3.3	0.8
Fluoranthene	202.26	0.0000067	0.0044	-3.2	0.8

a. ΔG_m = Gibbs free energy at minimum

b. PAH = Polycyclic Aromatic Hydrocarbons

Table 4. Contributions of Each of the Three Terms of the Thomson Equation to Total ΔG for Cluster Formation at the Minimum, x 10^{13} ergs

	ΔG_1 [a.]	ΔG_2 [a.]	ΔG_3 [a.]	ΔG_m [b.]
Pentachlorophenol	2.37	8.87	-20.58	-9.34
n-Butanol	6.75	4.06	-17.05	-6.23
i-Butanol	7.77	3.67	-17.08	-5.64
Phenol	5.45	6.58	-16.42	-4.39
Hexanal	5.95	4.46	-16.99	-6.58
Nonanal	4.20	5.63	-17.43	-7.60
Benzaldehyde	5.55	6.54	-17.90	-5.81
2-Butanone	8.02	3.69	-16.26	-4.55
Ethylacetate	6.65	3.60	-14.33	-4.08
2-Ethyoxyethylacetate	4.85	5.77	-17.18	-6.56
Nicotine	3.20	8.09	-19.81	-8.52
Chlordane	-0.24	9.96	-21.85	-12.13
Chlorpyrifos	0.64	10.55	-23.52	-12.33
Water	1.86	11.72	-17.87	-4.29

a. $\Delta G_1 = -nkT \ln S$; $\Delta G_2 = 4\pi r^2 \sigma$; $\Delta G_3 = \frac{q^2}{2}(1 - 1/\varepsilon)(1/r - 1/r_o)$

b. $\Delta G_m = \Delta G$ for cluster formation at the minimum

Table 5. Effect of Using Mean or Median Concentration on Values of ΔG_m

Compound	concentration, $\mu g/m^3$	$\Delta G \times 10^{13}$ ergs			
		ΔG_1	ΔG_2	ΔG_3	ΔG_m
n-Dodecane					
maximum	675	1.88	4.52	-9.97	-3.56
mean	9	2.55	4.06	-9.12	-2.51
p-Dichlorobenzene					
maximum	915	3.83	5.00	-12.15	-3.32
mean	2.8	5.35	4.44	-10.74	-0.94
Pentachlorophenol					
maximum	25	2.37	8.87	-20.58	-9.34
mean	5	3.68	7.65	-18.66	-7.33
Ethylacetate					
maximum	204	6.65	3.60	-14.33	-4.08
mean	6.8	7.51	3.40	-13.52	-2.61
Quinoline					
maximum	1.1	5.48	6.45	-16.09	-4.17
mean	0.022	5.96	5.75	-14.42	-2.70
Chlordane					
maximum	130	-0.24	9.96	-21.85	-12.14
mean	1.82	1.21	8.40	-20.13	-10.52
Chlorpyrifos					
maximum	87.7	0.64	10.55	-23.52	-12.33
mean	0.27	2.53	8.83	-21.42	-10.06

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
INFORMATION RESOURCES DEPARTMENT
BERKELEY, CALIFORNIA 94720