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Atmospheric ozone detection using a tin oxide solid state sensor

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ATMOSPHERIC OZONE DETECTION USING A TIN OXIDE SOLID STATE SENSOR

BY

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ABSTRACT

ATMOSPHERIC OZONE DETECTION USING A TIN OXIDE SOLID STATE SENSOR

By

Diana Lewis

University of New Hampshire, December, 2007

Solid-state gas sensors consist of a solid material, usually a metal oxide semiconductor, whose conductivity changes when gases adsorb onto its surface. The goal of this project is to create an ozone detection system using these sensors for use in autonomous environmental monitoring stations (such as weather balloons). To meet EPA specifications (less than 5% error), the sensor will have to have 1ppb resolution and be able to detect ozone in the range of 0-100ppb.

A sensor was purchased from MicroChemical Sensors (MiCS) for adaptation to this application but the stated range of application (10-1000ppb) was 10 times the desired resolution. The bulk of the effort on this thesis focused on calibrating the sensor at its lower detection limit. The sensor was tested for its response to ozone, as well as for crosstalk with H_2O, CO, NO_x, SO_2, and CO_2. The sensor generated a measurable response to ozone levels in the proposed measurement range, but had an equally strong response to humidity (it was insensitive to the other gases). The sensor response vs. humidity and ozone concentration were measured over a range of 0ppb to 150ppb ozone and 2.5% to 85% relative humidity. The results were fitted to a modified multiple-gas

Langmuir adsorption equation using an error minimization algorithm available in MATLAB. The data fit the modified Langmuir equation with an R^2 value of 0.9958.

The combined humidity and ozone equation was rearranged to obtain an equation for in-situ calculation of the ozone concentration. This was input into a LabVIEW program and tested against an ozone analyzer. The calibration held to within $\pm$ 2.8ppb, or $\pm$5% (with the exception of one extraneous data point), meeting EPA specifications.

CHAPTER I

INTRODUCTION

Project Motivation

Ozone at ground level is a pollutant caused by industrial activity. While it is not emitted directly from industrial sources, other products (NO_x and VOCs) combine in sunlight to produce it. At high levels, ozone causes respiratory health problems in humans and damages plant foliage, causing reduced crop yields and stunted forest growth [9]. Like other pollutants, ozone can be carried by the wind, making it important to track ozone and monitor it at rural sites as well as in urban areas.

The goal of this project was to design an ozone detection system using a solid state sensor. This detection system is to be used in autonomous environmental monitoring stations, both at ground level as well as in weather balloons. For this application they need to be small, lightweight, low power, and rugged. Solid state sensors fit all of these requirements, which is why they were chosen to try to fill this niche.

Because there are many solid state sensors available that can detect ozone, it was deemed unnecessary to design a new sensor, and one available model was chosen to adapt to this application. The sensor chosen was a MiCS 2610 ozone sensor, which was picked because it was one of only a couple of models found that were not already integrated into a sensing device. The detection range of this sensor is specified to be 10-1000ppb. AIRMAP data from Thompson Farm (where we will be testing our sensor) reported an average ozone concentration of 27 ppb for 2006, with a maximum hour-average of 93.3ppb and a minimum hour-average of 0.1ppb [1]. Note that our application requires monitoring ozone at or below the stated lower detection limit of the MiCS sensor most of the time and the bulk of our effort focused on extending the lower detection limit to ±1 ppb.

At this lower limit, one must worry about minor changes to the sensor, which could be caused by cross-talk from other gases, changes in ambient conditions (like pressure and temperature), as well as sensor aging. The goal of this project is to create a calibrated sensor system that will adjust for changes in these variables and output a reliable ozone measurement that meets EPA specifications (less than 5% error of reading).

History of Semiconductor Gas Sensors

The gas sensing properties of semiconductors were discovered in the 1950s when scientists working on p-n junctions discovered that they were sensitive to changes in the atmosphere. To them, this was considered a problem, which they fixed by isolating the junctions from the atmosphere. In 1962, Naoyoshi Taguchi began researching solid-state sensors with the hopes of competing with the catalytic bead sensor. In 1968, his company, Figaro Engineering, became the first to commercialize a solid-state gas sensor, called the Taguchi Gas Sensor (TGS) [2]. It was made of tin oxide and had poor selectivity towards various gas species. It was designed to sense high levels of combustible gases, just like the catalytic bead sensor. However, the new TGS had some advantages over its counterpart: it required less power and lasted about 10 years – compared to one to two years for the catalytic sensor. Four years later, IST (International

Sensor Technology) followed with a sensor that could detect hydrogen sulfide in the 0-10 ppm range. Nowadays, there are many types of SSGSs used for detecting numerous gas species. They are used in homes to detect ozone and CO, as well as in industry for flammable gas detection and oxygen detection for combustion processes.

There are many solid state sensors on the market today, most of which are used for detecting high levels of hazardous gases. Their low power consumption, low cost and small size lend themselves to many different markets. For instance, the automotive industry uses them to detect oxygen levels for combustion processes, as well as to monitor the hazardous gas levels in exhaust. They are starting to appear as personal warning devices that can be integrated into cell phones, or other electronic devices. Other applications include detecting hazardous levels of combustible gases, CO and ozone in the workplace/home.

The newest market for these sensors is environmental monitoring. This application requires sensors to be much more sensitive, selective and accurate than that of the previous generations. Instead of detecting hazardous levels of gases in the ppm range, they must detect low atmospheric concentrations in the ppb range to within 5%.

Basic Operation

Solid-state sensors consist of a solid semiconductor material, usually a metal oxide semiconductor, whose conductivity changes when gases adsorb onto its surface. To measure this conductivity change, the

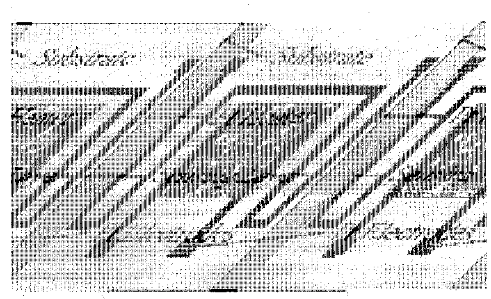


Figure 1: Thin film SSGS diagram

semiconductor is deposited as a thin film onto a substrate that has been patterned to create platinum electrodes and a heater. A voltage is applied to the sensor via the platinum electrodes to measure the conductivity and the heater is employed to elevate the temperature of the sensor to improve its gas sensing capabilities.

CHAPTER II

THEORY

Electronic structure of Tin Oxide

Tin oxide is a direct bandgap semiconductor with E_g=3.6 eV. The energy levels have been estimated using DFT (density functional theory) to solve the 3 dimensional Schrödinger equation [3] as shown in Figure 2.

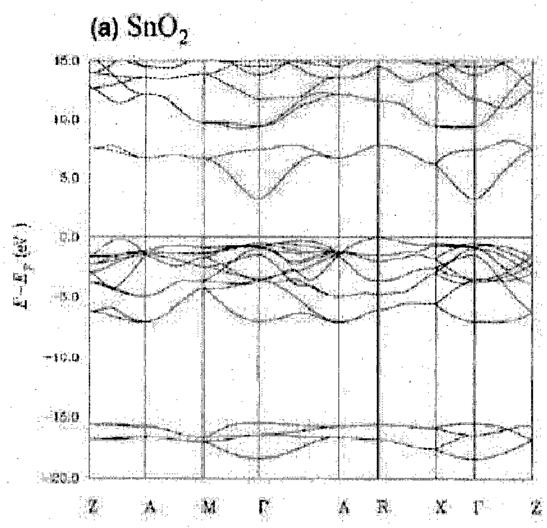


Figure 2: Band structure of SnO_2 [3]

It has a tetragonal structure with a=b=4.7374Å and c=3.1864Å. The chemical formula of the tin oxide used for sensing is SnO_{2-x}, where $x < 0.034$ (any more excess tin would precipitate out). Because there are oxygen vacancies, tin oxide is an n-type semiconductor.

The conductivity is therefore determined by the equation $\sigma = en\mu$, where e is the carrier charge, n is the charge carrier concentration, and μ is the mobility of the charge carrier. The carrier concentration is determined by the intrinsic carrier formula [5]

$$n_i = N_c \exp\left(\frac{E_f - E_c}{kT}\right) \quad \textbf{(1)}$$

where $N_c = 2(2\pi m_e^* kT/h^2)^{3/2}$ is the effective density of states at the conduction band edge, E_f is the Fermi energy, E_c the energy at the conduction band edge, m_e is the effective electron mass, k is the Boltzmann constant, h is Planck's constant, and T is the temperature. The mobility of the charge carrier is dependent upon the mean free path (ℓ) of the electrons in the semiconductor ($\mu = \ell/(m_e u)$, where u = mean speed). In the case of polycrystalline films, grain boundaries can have an effect on the mean free path if the grains are small enough ($1/\ell = 1/\ell_o + 1/\ell_g$, where ℓ_o is the original mean free path and ℓ_g is the mean free path in the grains), which would then lead to a decrease in conductivity ($\sigma_o/\sigma = 1.33\beta$, $\beta = (\ell/\ell_g)(R/1\text{-}R)$, R= probability of scattering), which in turn would adversely affect the sensitivity of the sensor. Also, the disordered nature of the grain boundaries causes a difference in the charge carrier density, resulting in a space charge region and creating a Schottky barrier that increases resistance. Because of these effects, one would think that an epitaxial film would be ideal. However, the dangling bonds at grain boundaries provide excellent sites for adsorption, which then lead to increased sensitivity. These two competing effects suggest that there is an optimum grain size for gas sensing. It is thought that the optimum grain radius is equal to the Debye length, which is the width of the space charge region. The Debye length is a function of the temperature and total carrier concentration (n_o) [5].

$$L_D = \left(\frac{\varepsilon_o kT}{n_o e^2} \right) \quad \textbf{(2)}$$

ε_o is the permittivity of free space. For T= 700K and n_o = 10^{22} /cm^3 (a typical concentration), the Debye length is 19.7nm.

Surface effects

The most common growth mode is along the (110) plane, which then becomes the surface. If the surface had no defects, it would have no sites for adsorption (except at the grain boundaries) [3]. In reality there will be a fair amount of surface defects, which would create a charged region and sites for adsorption just like the grain boundaries (note that the optimum film thickness would then be equal to the Debye length as well). For the purpose of calculations, it will be assumed that both effects contribute to the same surface potential.

Because the surface is in an oxygen-rich environment, oxygen will adsorb onto the Sn dangling bonds and dissociate into $2O^-$, drawing electrons up from the bulk and reducing the carrier concentration. The charge carrier density can be determined by the 1-D Poisson equation

$$\rho(z) = e[p(z) - n(z) + D^+(z) - A^-(z)] = \varepsilon_o \varepsilon_r \frac{d^2V}{dz^2} \quad \textbf{(3)}$$

where e is the charge, p and n are the densities of holes and electrons, respectively, D+ and A- are the densities of donors and acceptors, ε_o is the permittivity of free space, and ε_r is the dielectric constant of the material [3]. V is the potential due to band bending and z is the distance from the surface. Neglecting the hole concentration and assuming that the donor density is uniform ($D^+(z)=D^+$), the equation becomes

$$e[-n(z)+D^+]=\varepsilon_o\varepsilon_r\frac{d^2V}{dz^2} \qquad (4)$$

Assuming that $n(z)=D^+$ in the bulk and zero in the space charge region, one can integrate twice, define the bulk potential to be zero and substitute in the Debye length for the depth of the space charge region to yield

$$V_s=\frac{eD^+L_D^2}{2\varepsilon_o\varepsilon_r} \qquad (5)$$

Requiring that the space charge layer have an equal charge to the surface, $D^+L_D = N_s$, the equation becomes

$$V_s=\frac{eN_s^2}{2\varepsilon_o\varepsilon_r D^+} \qquad (6)$$

The sheet conductivity change due to the change in the surface charge is expressed as

$$\Delta\sigma_s=e\mu\int(n(z)-n_i)dz \qquad (7)$$

Integrating over the thickness of the semiconductor,

$\int_0^t n(z)dz=\int_0^{L_D}n(z)dz+\int_{L_D}^t n(z)dz=D^+t-D^+L_D$ (as defined above) and $\int_0^t n_i dz=D^+t$.

Substituting in for these expressions yields

$$\Delta\sigma_s=-e\mu D^+L_D \qquad (8)$$

Substituting in for D^+L_D yields the expression for the change in sheet conductivity due to surface charge.

$$\Delta\sigma_s=-e\mu N_s \qquad (9)$$

Adsorption

When a gas adsorbs onto the surface of a semiconductor, it will alter the sheet conductivity by changing the amount of charge on the surface

$$\Delta(\Delta\sigma_s) = -e\mu(\Delta N_s) \quad \textbf{(10)}$$

$\Delta\sigma_s$ is the change in sheet conductivity of the surface in relation to the bulk,

$$\Delta\sigma_s = \sigma_1 - \sigma_b \quad \textbf{(11)}$$

$\Delta(\Delta\sigma_s)$ is then

$$\Delta(\Delta\sigma_s) = \sigma_2 - \sigma_b - (\sigma_1 - \sigma_b) = \sigma_2 - \sigma_1 \quad \textbf{(12)}$$

The sheet conductivity is the inverse of the sheet resistivity, so the relationship between resistance and surface charge is as follows:

$$\frac{1}{\rho_{s2}} - \frac{1}{\rho_{s1}} = -e\mu(\Delta N_s) \quad \textbf{(13)}$$

Sheet resistivity has units of ohms/square; to convert to resistance one must multiply by a thickness, t, to get the resistivity, then use the resistance formula $R = \rho L/A_r$ (L is the length, A_r is the cross-sectional area of the surface layer) to get the resistance of the surface layer, R_s.

$$R_s = \frac{\rho_s t L}{A_r} \quad \textbf{(14)}$$

Substituting the resistance into the equation relating resistivity to surface charge yields

$$\frac{1}{R_{s2}} - \frac{1}{R_{s1}} = -A(\Delta N_s) \quad \textbf{(15)}$$

where $A = A_r e\mu/tL$.

Note that the change in resistance only occurs on the surface: outside of the space charge region, the bulk resistivity of the sensor does not change with surface charge, and therefore will not change when gases adsorb. The bulk resistance and surface resistance can be thought of as parallel resistors whose total resistance can be found by

$$\frac{1}{R} = \frac{1}{R_b} + \frac{1}{R_s} \tag{16}$$

where R_b is the bulk resistance and R is the total sensor resistance. The total sensor resistance is the measurable quantity. However, because we are measuring a change in resistance, the equations actually work out to a simple redefinition:

$$\frac{1}{R_{s2}} - \frac{1}{R_{s1}} = \left(\frac{1}{R_2} - \frac{1}{R_b}\right) - \left(\frac{1}{R_1} - \frac{1}{R_b}\right) = \frac{1}{R_2} - \frac{1}{R_1} \tag{17}$$

Using the total resistance and rearranging equation (15) to isolate R_2 yields

$$R_2 = \frac{1}{-A\Delta N_s + \frac{1}{R_1}} \tag{18}$$

The relationship between the change in surface charge and adsorbed gas species depends upon the nature of the adsorbed gas, and will be discussed individually in the next sections.

Oxygen Adsorption

As mentioned previously, in normal atmospheric conditions oxygen will adsorb onto the surface of the semiconductor, creating the space charge layer described in the previous section. This is proposed to occur in the following steps: oxygen is adsorbed onto an Sn dangling bond, capturing its electron. It then acquires another electron and dissociates into $2O^-$. At higher temperatures (above 400°C), a third step can occur, where the two adsorbed oxygens each capture another electron to become O^{2-} [4].

$$O_{2,gas} + e^- \leftrightarrow O^-_2$$

$$O_2^- + e^- \leftrightarrow 2O^-$$

$$2O^- + 2e^- \leftrightarrow 2O^{2-}$$

The adsorbed oxygen is thought to play a key role in the adsorption process of other gases, and has been assumed as the starting point for the surface charge and adsorption calculations.

Ozone Adsorption

The process of ozone adsorption is what causes our sensor resistance to change and allows us to detect its presence. There are a couple theories as to its adsorption process. The first theory is that ozone adsorbs onto a dangling Sn bond, then dissociates into an adsorbed oxygen and a gaseous O_2 molecule. The remaining oxygen then captures an electron from the bulk, becoming an O^- species on the surface.

$$O_{3,gas} + Sn_{lat} \leftrightarrow O_3\text{-}Sn_{lat}$$

$$O_3\text{-}Sn_{lat} \rightarrow O\text{-}Sn_{lat} + O_{2,gas}$$

The second theory is that ozone adsorbs onto an oxygen atom that is already on the surface, dissociates and releases O_2. The remaining two oxygens can then follow the same adsorption process as O_2.

$$O_{3,gas} + O_{lat} \rightarrow O_{2,gas} + O\text{-}O_{lat}$$

or

$$O_{3,gas} + O^-_{ads} \rightarrow O_{2,gas} + O^-_{2,ads}$$

In either case, ozone traps an electron, creating a surface charge and depleting the electrons in the space charge region. Because the mechanisms have the same result, it is impossible to determine which one is responsible for the sensor response, or whether it is a combination of both. From equation 18, the increased ΔN_s in turn increases the sensor resistance.

Water Adsorption

Water is an interfering gas. It also adsorbs and causes a resistance change in the sensor. Infrared spectroscopy data indicates that there are three types of hydroxyl groups on the surface in the presence of water: a chemisorbed hydroxyl group, M-OH; a rooted hydroxyl group, O_{lat}-H; and hydrogen bridged hydroxyl groups [4].

The theories for water adsorption include the following process: Sn and O lattice atoms dissociate a water molecule into two OH groups.

$$H_2O_{gas} + Sn_{lat} + O_{lat} \rightarrow Sn_{lat}\text{-}OH + O_{lat}\text{-}H^+ + e^-$$

The oxygen in the lattice steals the electron from the hydrogen, reducing the surface charge and donating an electron to the bulk.

A similar theory is that water dissociates and adsorbs onto a tin atom and an adsorbed O^-, creating two hydroxyl groups attached to lattice tin atoms. The electron from the hydrogen would then be redistributed to the bulk. Another theory is that water uses up an adsorption site for oxygen, which then can't steal an electron from the bulk.

In any case, one electron is released to the bulk for each adsorbed water molecule, even though it dissociates into two adsorbed species. This means that ΔN_s is negative for adsorbed water, and the resistance will drop as ΔN_s increases.

Comments

It is important to note that for any combination of a theory for ozone adsorption and a theory for water adsorption, water blocks at least one site for ozone adsorption, and vice versa. This is important as it influences the choice of an adsorption equation to model the system.

Adsorption Isotherms

Langmuir Adsorption

When a surface is put in contact with a gas, gas molecules will adsorb onto the surface until equilibrium is reached. There are a few different theories on adsorption modes: the most widely known is the Langmuir adsorption process. Langmuir's theory assumes the following: adsorption takes place at a constant number of isolated adsorption sites that do not interact with one another and that have the same adsorption energy. The following equation for the rate of adsorption is based on these assumptions:

$$\frac{dN}{dt} = \alpha P(N_t - N) - \beta N \quad (19)$$

where the probability of adsorption is $\alpha = \kappa S/\sqrt{(2\pi MkT)}$ and the probability of desorption is $\beta = \nu \exp(-E_d/kT)$. P is the partial pressure of the gas, κ is the sticking probability, S the fraction of molecules possessing the energy to adsorb [6], M the mass of the adsorbed molecule, ν the desorption coefficient and E_d the energy of desorption. N_t is the total number of adsorption sites per unit area and N is the number of adsorbed species per unit area [7]. Notice that both the adsorption and desorption terms are temperature dependent, and that the desorption term increases faster than the adsorption term. This means that as the temperature increases, the system comes to equilibrium faster (and has a lower coverage, but this decrease in sensitivity is a tradeoff). Generally speaking, the sensor's maximum stable temperature (before beginning to anneal) is the upper limit for the operation temperature. For tin oxide sensors, the temperature at which grain growth begins to occur is 450°C. The operating temperature of the MiCS sensors is approximately 430°C. The following is a simulated plot of adsorption coverage vs. time at different temperatures for an ozone concentration of 50ppb. The red line is the normal operating temperature of the sensor. Notice that an increase of just 20°C causes the time constant to drop from about 75 seconds down to about 45 seconds (dashed line to dashdot line), and decreases the fractional surface coverage from 0.63 to 0.37.

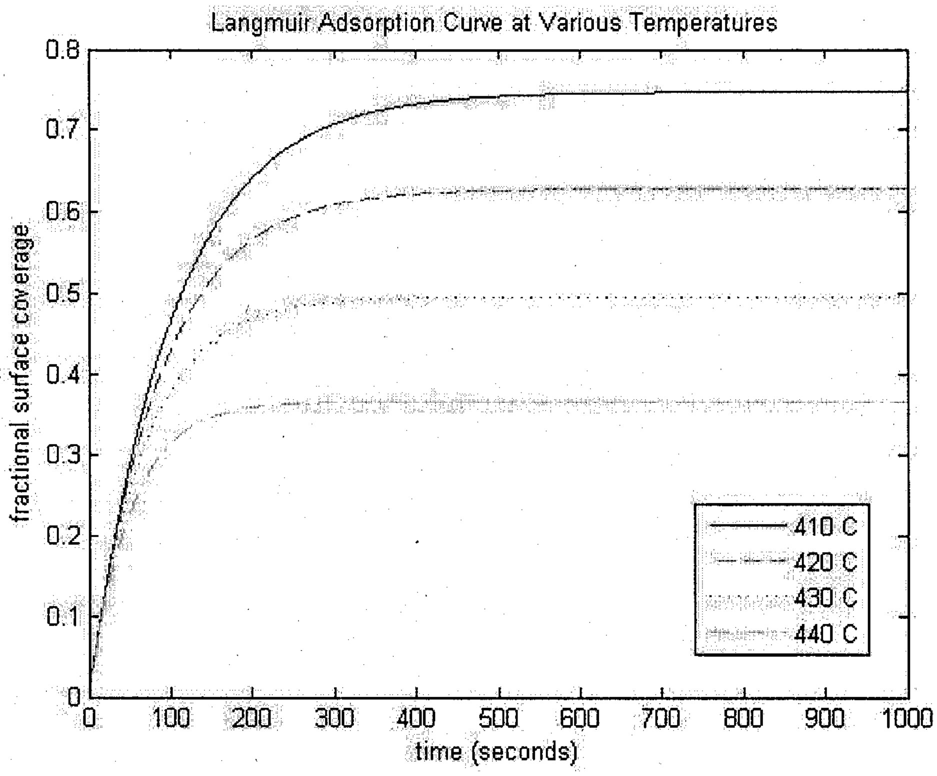


Figure 3: Adsorption curves at various temperatures, simulated using the adsorption rate equation

The plot was generated using the following values for the constants:

m = 48/(6.02e23*1000) kg/molecule
Ed = 2.25*1.602e-19 J
ν = 1e14 Hz
κS = 1e-22 kg-m

The mass of an adsorbed ozone molecule is 48 g/mol. This value was converted to kg/molecule for unit matching. The energy of desorption, Ed, was assumed to be 2.25 eV/molecule (converted to joules for unit matching) because values found for energies of desorption for other systems were in the range of 2-3 eV/molecule (no value could be found for the energy of desorption of ozone on tin oxide). ν was defined as 10^{14} Hz which is a typical number for a lattice vibration. κS was chosen to obtain curves in the range of what was seen in our lab experiments. See appendix for complete MATLAB code.

To find the equilibrium surface coverage, the rate equation (19) is integrated:

$$N(t) = \left(\frac{N_t}{1+\frac{b}{P}} \right) (1-\exp(-at)) \qquad \textbf{(20)}$$

where a = $\alpha P+ \beta$ and b = β/α. At long times, the exponential term drops out and the equation becomes the Langmuir isotherm

$$N = \frac{N_t}{1+\frac{b}{P}} \qquad \textbf{(21)}$$

Modifications to Langmuir's Isotherm

Multiple adsorbing gases

If there are more than one type of gas adsorbing onto the surface that both use the same adsorption sites, Langmuir's isotherm is adjusted to [7]

$$N = N_t \left(\frac{\frac{P_i}{b_i}}{1+\Sigma \frac{P_i}{b_i}} \right) \qquad \textbf{(22)}$$

Note that this equation can be used to model the adsorption of humidity and ozone if one can assume that they use the same adsorption sites.

Activated adsorption

In normal adsorption, the sticking probability κ is assumed to be constant. If the adsorption is thermally activated, the sticking probability becomes [7]

$$\kappa = C\exp\left(\frac{-E_a}{kT}\right). \quad \textbf{(23)}$$

Note that for our setup at constant temperature, the temperature dependence of the sticking probability is irrelevant.

Other Types of Adsorption

Because Langmuir's adsorption equation contains simplifying assumptions, experimental results do not always correspond to the theoretical equation. If one modifies the theory to allow for a heat of adsorption that varies with surface coverage, one obtains the Fruendlich isotherm:

$$N = CP^n \quad \textbf{(24)}$$

where C and n are constant and n<1 [7]. The Freundlich isotherm is generally used over small coverage ranges.

Another type of adsorption is the logarithmic isotherm [7], which follows the equation

$$N = C\ln\left(\frac{P}{P_o}\right) \quad \textbf{(25)}$$

There are many other types of adsorption equations as well. Some are attempts to fit the data, while others are theoretical models like the Langmuir adsorption model. All have value; however, we will be using the Langmuir model to fit the data.

Equation Relating Partial Pressure to Sensor Resistance

To find a relationship between the sensor resistance and the gas pressure, one must relate the amount of gas adsorbed with the change in sensor resistance. This can be achieved by combining an adsorption isotherm with equation (18) (the equation relating resistance to surface charge). The adsorption isotherm chosen to model the system is the Langmuir isotherm for multiple gases (equation 22).

For ozone adsorption, each ozone molecule that is adsorbed captures an electron, increasing N_s. Therefore, the relationship between adsorbed molecules and the change in surface charge is

$$N = N_{oz} = \Delta N_{s,oz} \tag{26}$$

For water adsorption, the opposite is true. Each water molecule that is adsorbed releases an electron from the surface, decreasing N_s. Therefore, the number of adsorbed molecules is the negative of the change in N_s.

$$N = N_w = -\Delta N_w \tag{27}$$

The total change in charges on the surface is then

$$\Delta N_s = \Delta N_{s,oz} + \Delta N_{s,w} = N_{oz} - N_w \tag{28}$$

Substituting this relationship into equation (18) produces

$$R_2 = \frac{1}{-A(N_{oz} - N_w) + \frac{1}{R_1}} \tag{29}$$

The Langmuir isotherm for multiple gases (equation 22) gives us

$$N_{oz} = N_t\left(\frac{\frac{P_{oz}}{b_{oz}}}{1+\frac{P_{oz}}{b_{oz}}+\frac{P_w}{b_w}}\right) \quad \textbf{and} \quad N_w = N_t\left(\frac{\frac{P_w}{b_w}}{1+\frac{P_{oz}}{b_{oz}}+\frac{P_w}{b_w}}\right) \tag{30,31}$$

Substituting into equation (29) yields the final equation for the resistance of the sensor as a function of ozone and water concentrations in the atmosphere.

$$R_2 = \frac{1}{-AN_t\left(\frac{\frac{P_{oz}}{b_{oz}}-\frac{P_w}{b_w}}{1+\frac{P_{oz}}{b_{oz}}+\frac{P_w}{b_w}}\right)+\frac{1}{R_1}} \tag{32}$$

A variation of this equation is to allow a different total number of adsorbed molecules (N_t) for ozone and for water.

$$R_2 = \frac{1}{-A\left(\frac{N_{t,oz}\frac{P_{oz}}{b_{oz}}-N_{t,w}\frac{P_w}{b_w}}{1+\frac{P_{oz}}{b_{oz}}+\frac{P_w}{b_w}}\right)+\frac{1}{R_1}} \tag{33}$$

This is a simple assumption that allows an extra variable to be inserted into the resistance equation, but implies that the surface sites are not independent of each other (a fundamental assumption of the Langmuir isotherm). A possible reason for the difference in total adsorption sites that wouldn't disrupt the other Langmuir assumptions would be that the ozone sites and the water sites are independent of each other, but that the water adsorption blocks the adjacent sites for ozone adsorption, and vice versa.

Another possibility is that the number of donated electrons is more or less than predicted: if the ozone molecules used up more than one electron by adsorbing, or if the water molecules donated more than one electron, the equation relating resistance change to the number of adsorbed molecules (equations 26 and 27) would need a coefficient that might be different for water and ozone. This would result in the same insertion of the extra variable (the definition of $N_{t,oz}$ and $N_{t,w}$ would then include the coefficients).

CHAPTER III

EXPERIMENTAL METHODS

Sensors

The ozone sensors were purchased from MiCS (Microchemical Sensors). The actual sensor is enclosed in a metal casing, with a metal screen over the top of the sensor. It has a detection range of 10-1000ppb and can operate between -40°C to 70°C. (See Appendix D for complete specifications.)

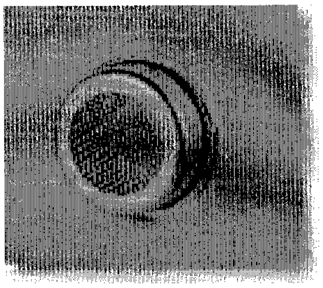


Figure 4: MiCS sensor

We dissected the sensors to determine their configuration. The screen was removed to reveal a 2mm square chip attached with glue to the casing and wire-bonded to the legs of the casing. Three of the legs are isolated from the rest of the metal by being suspended in epoxy: the fourth is the heater ground and is soldered directly to the casing. The sensor material and electrodes are located in the center of the sensor. In figure 5 the sensor electrodes are the horizontal yellow bars leading to the dark brown; the vertical ones lead to the pink heating element, which runs underneath the sensor (gold).

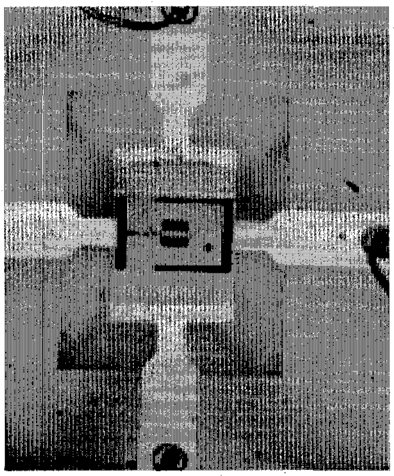


Figure 5: Close-up of sensor chip.

The center of the chip appears to be very thin; this was confirmed when the chip was removed from the casing. Figure 6 shows the chip flipped upside-down. The four blobs are the glue that held the sensor in the casing. The thin center is what allows the heater to run with such low power.

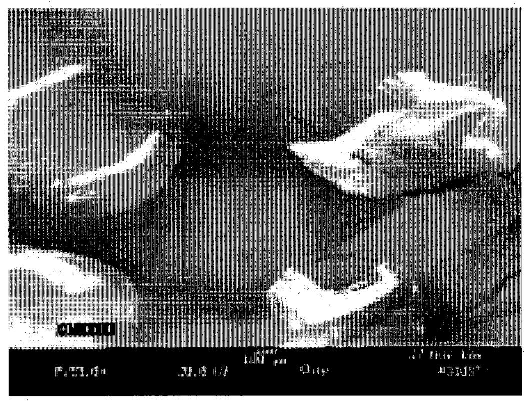


Figure 6: SEM image of reverse of chip

The chip was taken to the scanning electron microscope (SEM) for further analysis. The close-up SEM picture is of half of the sensor section, with the heater running vertically and the sensor electrodes running horizontally. The picture shows four distinct regions. The lightest sections are the electrode and heater; the next lightest section is in between the electrodes and is the sensing material; the third section is between the heater electrode and the sensor, and the darkest section in the upper right of the picture is the substrate.

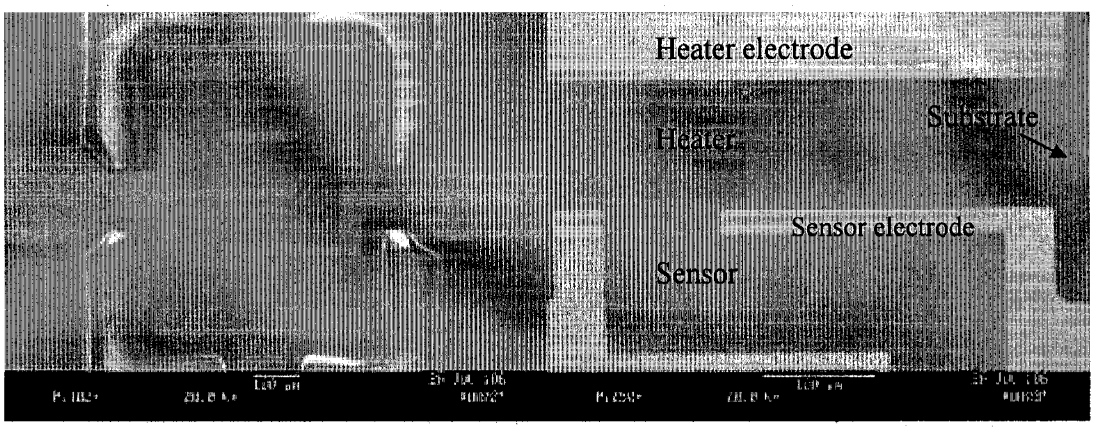


Figure 7: Left – SEM image corresponding to Figure 6. Right – close up of upper half of sensing area, showing four distinct layers.

The close-up picture was analyzed using electron dispersion spectroscopy (EDS) to determine the composition of the four sections. The electrodes are platinum, the sensor is tin oxide, and the substrate is silicon. The heater appears to be silicon dioxide, but this is an insulator so it is suspected that this layer was deposited on top of the heater to isolate it from the sensor. We were not able to determine the composition of the heater.

Note that the sensor electrodes have a finger extending into the middle of the sensor. This is to reduce the path length between the electrodes and thus reduce the sensor resistance to a reasonable value. Also, in the microscope image, the very center of the chip appears to have a distinct square that is not immediately discernable in the SEM picture. The EDS from the SEM does not detect any difference in material of the square vs. the surrounding area. Upon further inspection of the reverse side of the sensor, the square appears to be a section that is missing the heater layer. It is not known why the section would be missing.

A sensor was cracked in half to obtain a cross-section. The center of the chip is shown in the picture below. The thickness is approximately 3.25μm. There appear to be three distinct regions; the top layer is assumed to be the sensor layer, which is approximately 1.14μm thick. The other two layers are not identifiable using EDS maps. Point EDS may have worked but it was not attempted.. We suspect that the white layer is the SiO_2 chip and the middle layer is the heater.

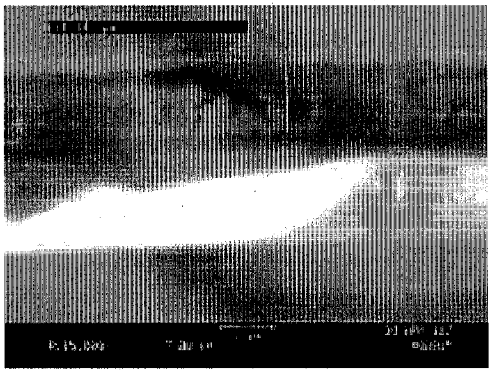


Figure 8: Cross section of chip. Line indicates thickness of top layer (thought to be the sensing layer).

Preliminary Tests

The sensors were originally set up according to the manufacturer specifications. A simple circuit board was created to deliver a constant 2.35 V to the heater. It consisted of voltage inputs to the heater and sensor and a load resistor in series with the sensor. The voltage across the sensor was measured, which then allowed for the sensor resistance to be calculated using the voltage divider rule.

$$R_s = \frac{R_L V_s}{V_{in} - V_s} \tag{34}$$

To measure the effects of ambient conditions, the circuit board also included a temperature sensor (National Semiconductor LM60), humidity sensor (Honeywell HIH-4000) and pressure sensor (Motorola MPX4250A). The sensor was encased in a glass tube and a pump was installed to ensure a constant flow rate across the sensor. An ozone generator was attached to the sensor setup using Teflon tubing and Swagelok fittings, and a bubbler was installed to humidify the air after the ozone generator. The bubbler was heated using heat tape to obtain the highest humidity possible.

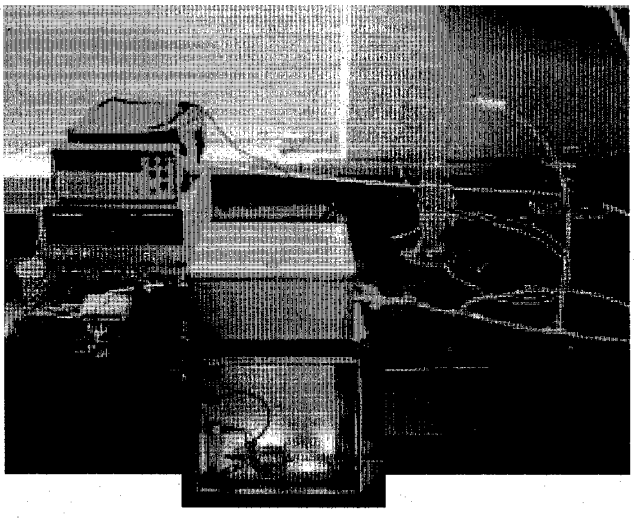


Figure 9: Preliminary setup

We determined that the sensor was sensitive enough to detect low levels of ozone, but was also sensitive to humidity. Sensor response to other gases was tested by replacing the ozone generator with an Environix series 100 mixing box and gas cylinders containing fixed concentrations of various other gases. The sensor did not respond to atmospheric levels of CO, NO_x, SO_2, or CO_2.

The sensor response using this setup was not repeatable. After communicating with MiCS, it was determined that the problem was heater aging – the heater resistance changes with time, which caused the sensor's temperature to drop if a constant voltage is applied (recall that a temperature change causes a change in the amount of gas adsorbed). The company suggested trying to keep the heater power constant instead of applying a constant voltage.

Modified Sensor Setup

Heater Control

While the company suggested that the sensor be run at constant power, we decided to try to keep the sensor temperature constant because constant power would not accommodate changes in ambient conditions (such as flow rate and temperature). The sensor chip did not have a temperature sensor, but the heater's resistance changed linearly with temperature and was calibrated to be used as its own RTD. Unfortunately, the heater's resistance increase over time was unpredictable and there was no way to recalibrate the RTD in-situ. (It was originally thought that the heater aging would only cause the intercept of the RTD calibration to change, but not the slope. If this were true, one could determine the intercept by measuring the resistance at an almost-zero voltage and correlating it with room temperature to obtain a data point. That coupled with the previously acquired slope would yield an updated calibration curve.) After the constant temperature experiment was unsuccessful, we decided to heed the company's suggestion and use a constant power setup. In future work, we will account for changes in ambient temperature using standard heat transfer considerations.

Test setup

The test sensor was encased in a glass tube along with the temperature and humidity sensors and attached to the new board. To ensure the sensors were sealed inside the glass tube, their bases were wrapped in Teflon tape and friction-fit into pre-cut holes in the glass tube. The whole tube was then wrapped in Teflon tape and covered with heat-shrink tubing. The glass tube is connected via tubing to a pump and a pressure sensor on the reverse side of the circuit board. The pump is situated after the sensor and pulls the test air through the glass tube, thereby not disrupting the ozone signal. The pump was disconnected during lab tests to allow the flow rate to be controlled externally.

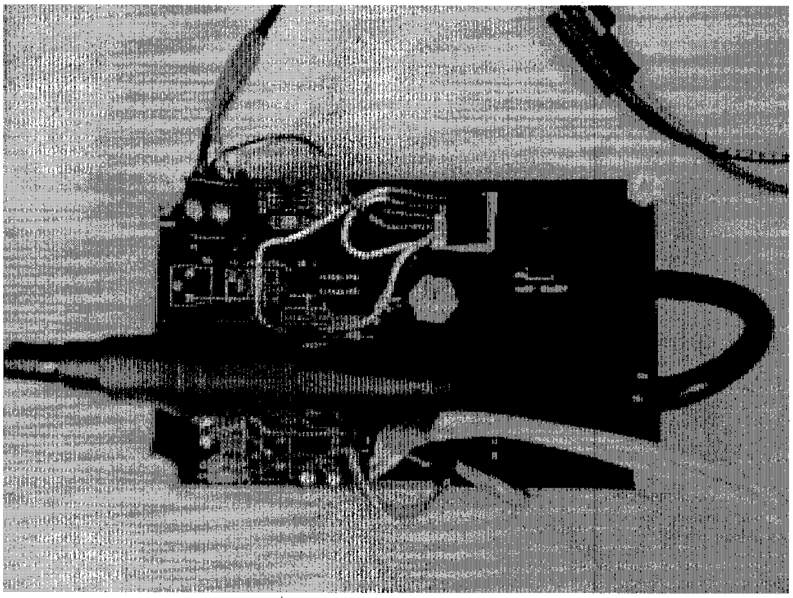


Figure 10: Ozone board.

This setup was placed inside a metal box with a Teflon tube leading from the inlet of the glass tube to the outside of the box. The metal box should provide a stable, protected environment for field-testing. For the purpose of lab experiments and calibration, test air is supplied via Teflon tubing by an ozone generator (Environix series 100 mixing box), which draws in zero air from a building supply. A loop was built into the tubing to divert some of the air through a bubbler for humidification. Shortly after the humidity loop, another split leads to a calibrated ozone analyzer (Thermo Electron 49C) that verifies the ozone concentration.

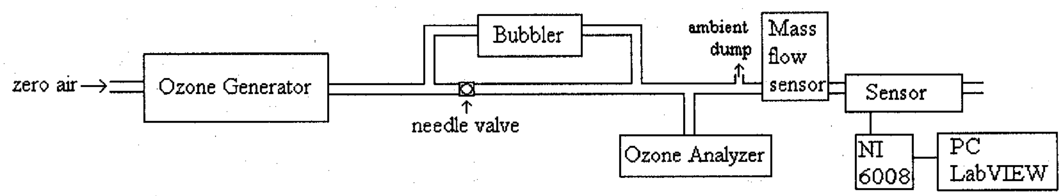


Figure 11: Test setup

A mass flow sensor (MKS mass-flo meter) was installed between an ambient dump and the metal box to monitor the flow rate. The circuit board was powered with a BK-precision 1760A DC power supply and connected to the computer via a National Instruments USB-6008. A LabVIEW program was developed to control the heater power using a PID controller, as well as to read out the sensor signals. (See Appendix B)

Testing

The sensor was powered at 80 mW and the flow rate controlled at 0.5 L/min. The flow rate was chosen because the pump was thought to pull at that rate. The power level was chosen to adhere to manufacturer specifications. This power level should cause the sensor to operate at approximately 430°C in still air. (Under test conditions, the flow rate of 0.5 L/min will cause the temperature to decrease by a small amount – approximately 10°C - due to an increase in the heat transfer coefficient, but the change was ignored for this set of testing. In future work, the power will be increased to compensate for the cooling effect of the flow.)

Two protocols were used to obtain the data to calibrate the humidity and ozone response of the sensor. In all methods, we waited 15 minutes after each change to achieve steady state before recording the data. In the first method, the humidity was set to a constant value and the ozone concentration varied. The ozone concentration was stepped from zero to 200ppb in 50ppb increments, and then set back to zero before changing the humidity. The ozone concentration was varied low to high because the time to steady state when increasing the ozone concentration was twice as fast (approximately 800 seconds vs 1600 seconds) than the time to steady state when decreasing the ozone concentration. (The time constants were 60s vs 90s).

The second test method was to set an ozone concentration between zero and 200, then vary the humidity. The humidity was varied in small, uneven steps (2%-10%) between approximately 20% and 85% RH. This method was more common because it was easier to keep the ozone level constant than it was the humidity.

The LabVIEW program recorded 10-second averages of humidity, ambient temperature, pressure, heater power, and sensor resistance. The ozone concentration was recorded by hand directly from the ozone analyzer. Throughout testing, the ambient temperature stayed between 25°C and 28°C, and the pressure was between 753 Torr and 763 Torr (100.3 kPa and 101.7 kPa).

CHAPTER IV

RESULTS

Preliminary tests

From the preliminary tests, it was determined that the sensor was not responsive to atmospheric concentrations of CO, NO_x, SO_2, or CO_2. It was determined that the sensor needed a humidity correction and that a constant heater power approach would be best for long-term repeatability of the sensor.

R_s vs Ozone Curves

To obtain a sensor resistance vs. ozone curve, the steady state resistance at each ozone concentration was averaged over 200 seconds and plotted. Data was gathered at multiple humidities. The results are plotted in Figure 12 for 3.5%RH. Note that the standard deviation for each point is plotted, but is too small to be seen. The maximum standard deviation for any point was 782 ohms (less than 0.3 ppb) at 50ppb. See Appendix F for complete data.

Notice that the sensor response has its steepest slope below 100ppb – an average of about 3000 ohms/ppb. This indicates that the sensor is most responsive in the low ppb range, and that small changes in ozone concentration result in easily measurable resistance changes.

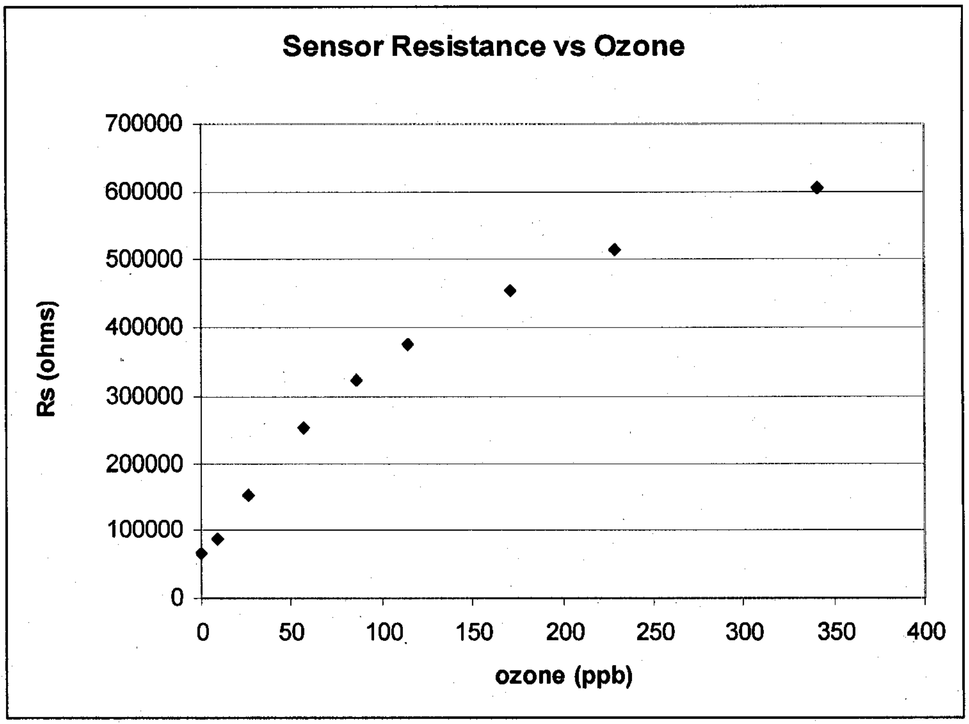


Figure 12: Steady-state ozone data at 3.5% relative humidity

R_s vs Humidity

The sensor resistance vs. humidity was analyzed and plotted in a similar manner. For the humidity data, the transient response data was excluded and the remaining data points plotted without averaging. The curve for zero ozone is shown in Figure 13. Notice that the sensor resistance decreases with increased humidity, while the sensor resistance increased for ozone. This is the expected result of the electron transfer that occurs during adsorption (see Adsorption section).

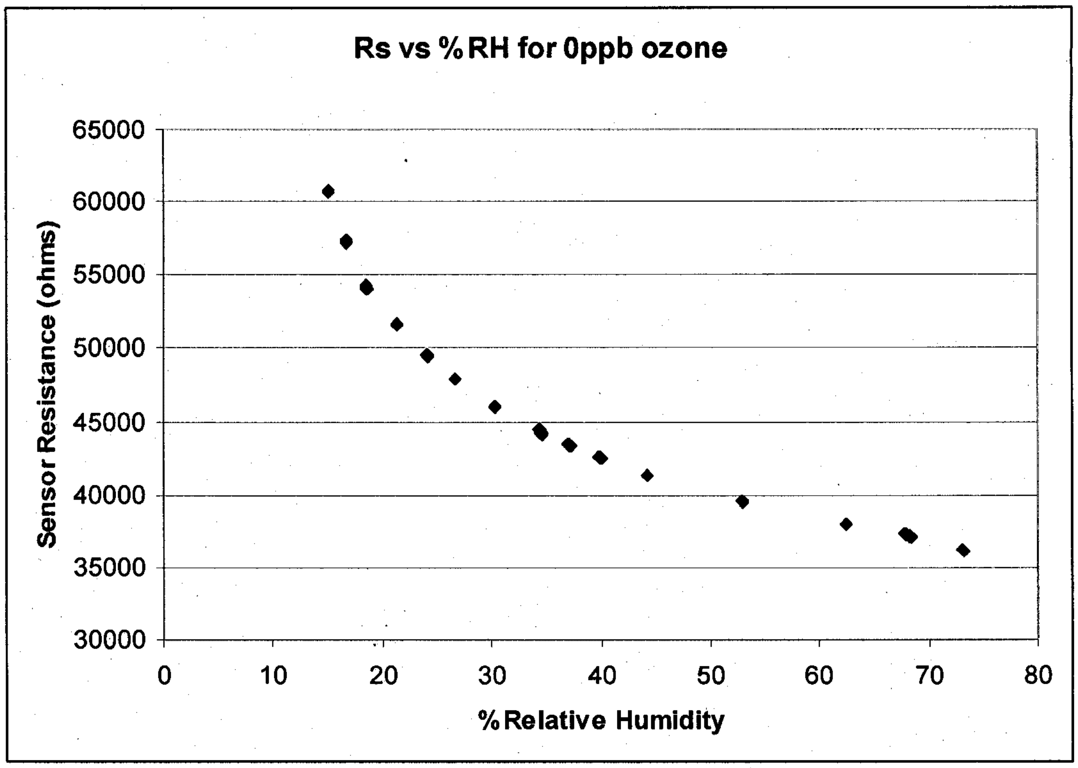


Figure 13: Steady-state humidity data at zero ozone concentration

Repeatability

The sensor response to humidity and ozone were tested on multiple days to determine if the results were repeatable. After excluding the transient response data, the remaining steady-state data points were plotted without averaging. As shown in Figure 14, the sensor does respond in a repeatable fashion, although there seems to be some error between the different days, especially at higher ozone concentrations (maximum difference was 11468 ohms - about 4ppb - at 100ppb and 47% RH). The error may be caused by changes in the ambient conditions, such as temperature or pressure. Testing has not yet been performed to determine if these factors are the cause. Note that the data set for 24.5ppb on 6-20 exhibited a spike in the sensor resistance around 57% RH. It is unclear why this happened; the acquired data doesn't show any difference in the ambient conditions or in the power to the sensor.

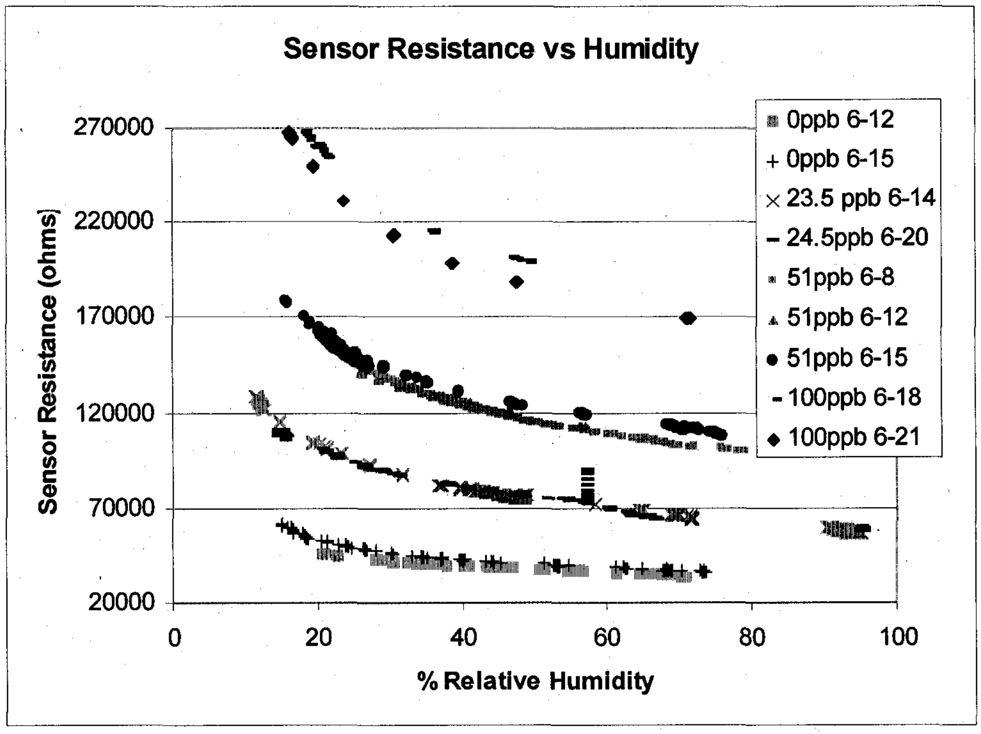


Figure 14: Compiled data from multiple days, humidities, and ozone concentrations

CHAPTER V

DISCUSSION

Langmuir Fit of Ozone Data

The Rs vs Ozone curve at low humidity was tested for fit to the Langmuir isotherm. The reciprocal of the change in resistance was plotted against the reciprocal of the pressure (assume zero humidity and rearrange equation 33) to determine if the data fit to the Langmuir isotherm. R_1 (the sensor resistance at zero ozone) was 63300 ohms.

$$\left(\frac{1}{\frac{1}{R_2} - \frac{1}{R_1}} \right) = \frac{b}{\frac{1}{R_t} - \frac{1}{R_1}} \left(\frac{1}{P} \right) + \frac{1}{\frac{1}{R_t} - \frac{1}{R_1}} \tag{35}$$

If the resulting curve is linear, the data fits a Langmuir isotherm. R_t, the saturation resistance, can be determined from the intercept, and b can be determined from the slope. Figure 15 shows all of the data plotted with a linear fit. The ozone concentrations were converted from ppb to kPa using an average value of pressure in the test chamber (101.64kPa) using the following formula. X is the ozone concentration in ppb.

$$P_{oz} = P_{chamber} X * 10^{-9} \tag{36}$$

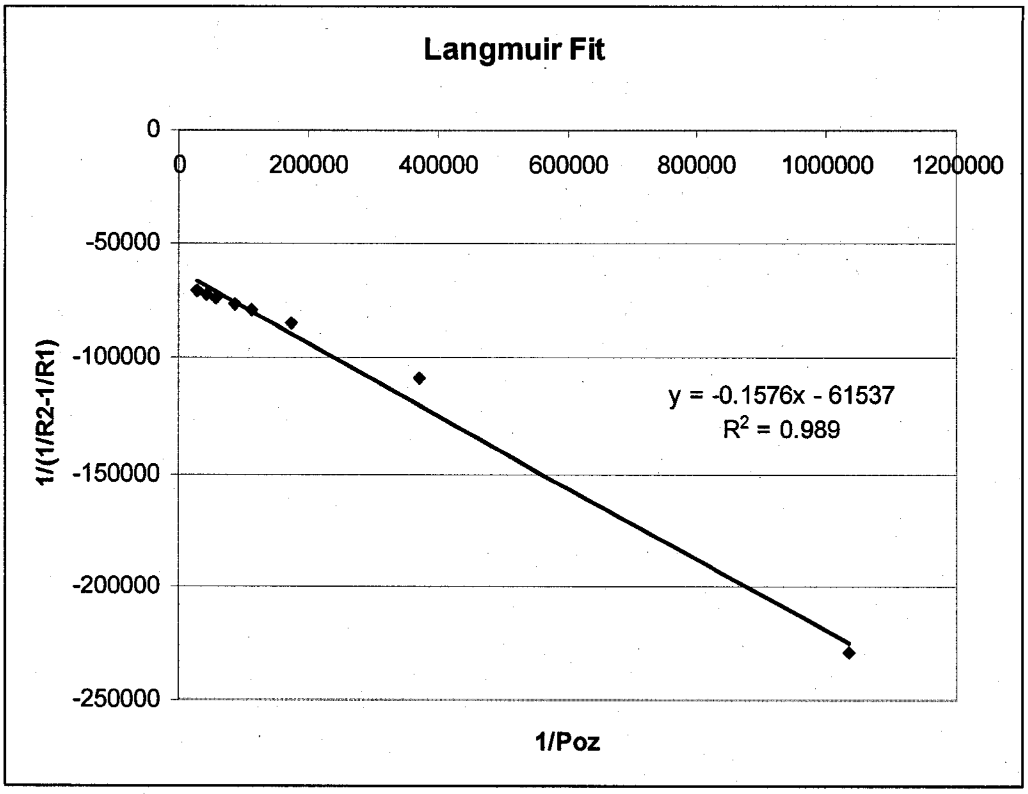


Figure 15: Langmuir fit for steady-state ozone data

The plot above shows a correlation to the Langmuir isotherm, but isn't perfect. However, the precision of the ozone analyzer is ± 1ppb, which means that the 9ppb data point (lower rightmost point on the plot) has 11% uncertainty and should not be included in the fit. When this value was excluded, the rest of the data fit a Langmuir isotherm with an R^2 value of 0.9958 (see figure 16). The calculated saturation resistance from this plot was 1187252 ohms. The calculated b value was 1.642e-6 kPa.

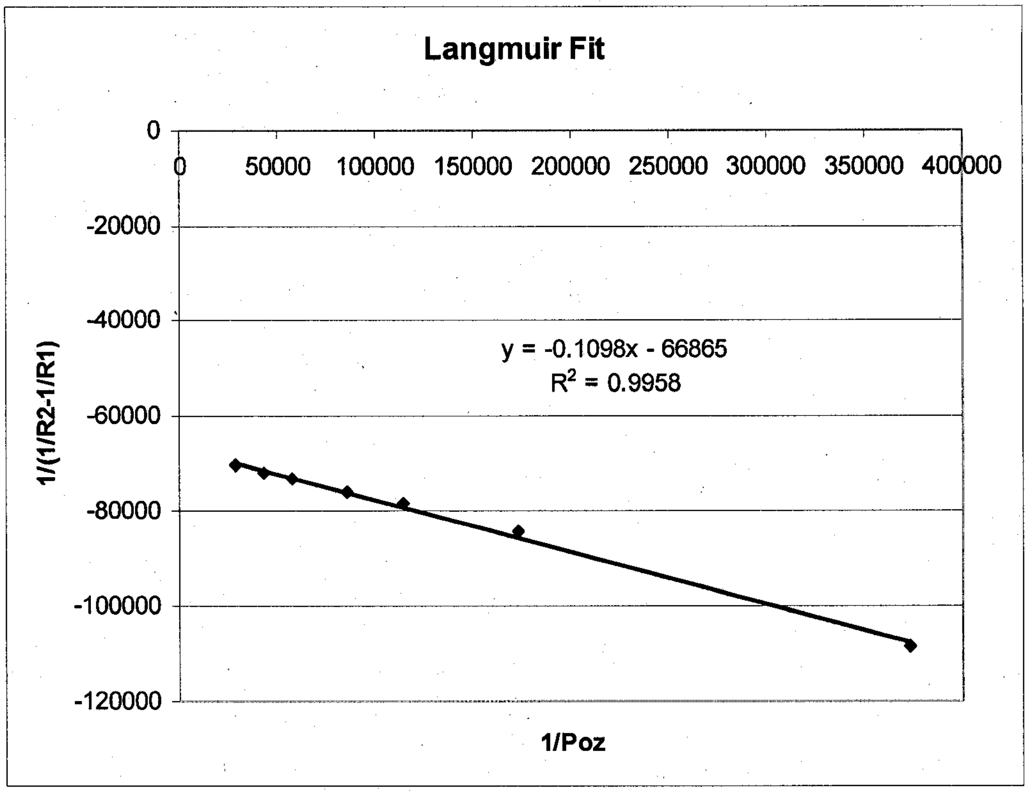


Figure 16: Langmuir fit for steady-state ozone data, excluding the 9ppb data point

Because of this fit of the data, the sensor response to ozone was characterized using a Langmuir isotherm. A MATLAB program was used to determine the best fit constants in the rate equation (equation 20), $N(t) = [N_t/(1+b/P)][1-\exp(-at)]$. Recall that $b = \beta/\alpha$, and $a = \alpha P + \beta$. b was determined from Figure 16, so the ratio of β to α is fixed. A response curve from 0-40ppb at 2.5%RH was chosen to try to fit the model (this data was chosen so that the humidity influence could be neglected). Our MATLAB program ***fitlang.m*** (see Appendix A) was used to find α (and β) by fitting the rate equation to this curve. It was accomplished by inputting the b value obtained from a plot like Figure 16 above (note that the file oz_0RH.xls was used for this data: the b value was 2.3623e-6 kPa) and finding the value of alpha that produced the minimum of the sum of squared errors (sse) using the MATLAB function fminsearch (fminsearch uses an initial guess to find the local minimum of the input function - in this case sse). The results are shown in Figure 17: the lower set of crosses is the data used to fit the model, and the line is the fitted curve. The value obtained for α was 2939.6 $(kPa\text{-}s)^{-1}$ and β was then determined by the equation $b = \beta / \alpha$ to be 0.00694 s^{-1}. (The program was then re-run to obtain beta first and then extract alpha from the relation – they were equivalent.) Another program (see Apendix A for ***fit_ab.m***) was written to check this calculation, which plotted the data for 40-80ppb (upper set of crosses) and then plotted the curve for that data using the obtained α and β.

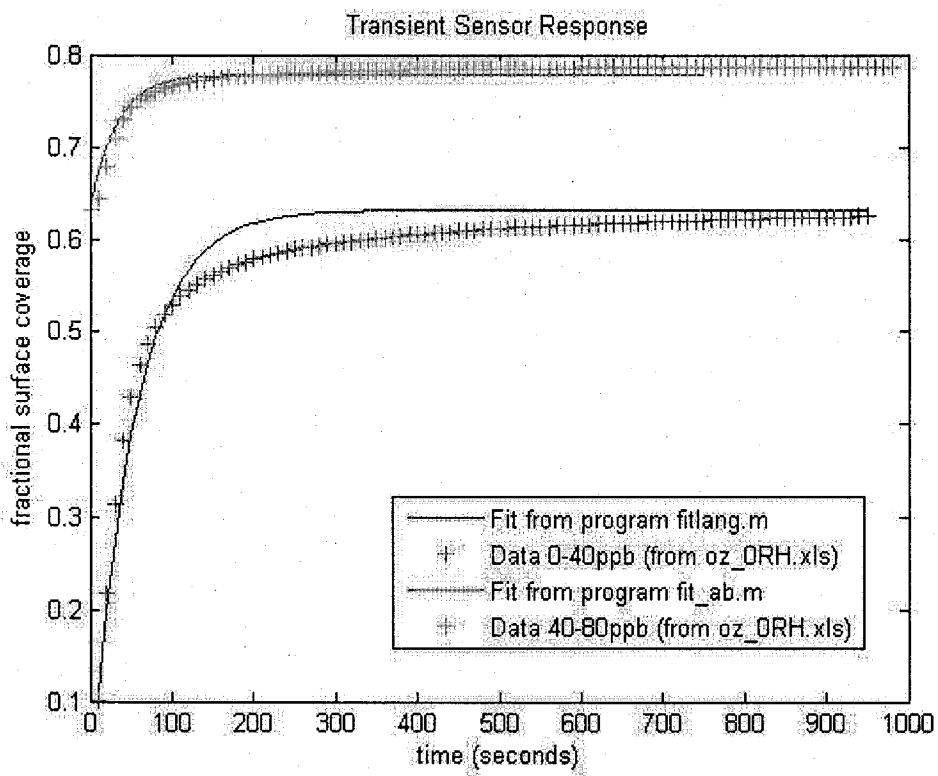


Figure 17: Transient Sensor Response. Sensor data (lower set of crosses) plotted against fitted curve from MATLAB program. The upper data set was used to check the fit of the curve at a different ozone concentration change.

One may notice that the shapes of the data and fitted curve differ. There are a couple of reasons why this could occur: first, the time to change the gas concentration in the chamber could be equivalent to the sensor response, and second, the sensor could have a non-Langmuir transient response to adsorbed gases. In idealized slug flow, the chamber would be completely flushed in 1.24 seconds (see Appendix E for calculation), which would indicate that the response seen is due to the sensor itself. However, the sensor is inside a screened-off volume of air (refer to figure 4) that may take longer to reach equilibrium with the test chamber, which means that the difference could be due to the testing environment. If the response is due to the adsorption behavior of the sensor, it could mean that there are multiple steps to the adsorption process, or that the Langmuir adsorption equation is not a perfect model for this system.

Note that β and α each contain two physical constants ($\alpha = \kappa S/\sqrt{(2\pi MkT)}$ and $\beta=\nu\exp(-E_d/kT)$). If the sensor was run at two different known temperatures, ν, E_d, and κS could be derived (and the mass, M, verified). However, at the present time, the temperature of the sensor cannot be accurately determined. (We were unsuccessful in our attempt to obtain the temperature by using the heater as an RTD - see Heater Control section, p24. We then attempted to obtain the temperature with an infrared sensor, but the spot size encompassed the tin oxide as well as the platinum electrodes and some of the silicon chip – an emissivity was needed to calibrate the infrared sensor, and it was impossible to determine the correct value.)

Langmuir Fit of Humidity Data

The sensor response to humidity was also tested for fit to the Langmuir isotherm. Because there was no data at low relative humidities, the starting value of relative humidity was taken to be 15%. That is, the change in resistance is taken to be the change in resistance from the resistance at 15% RH, and the change in relative humidity is RH - 15%.

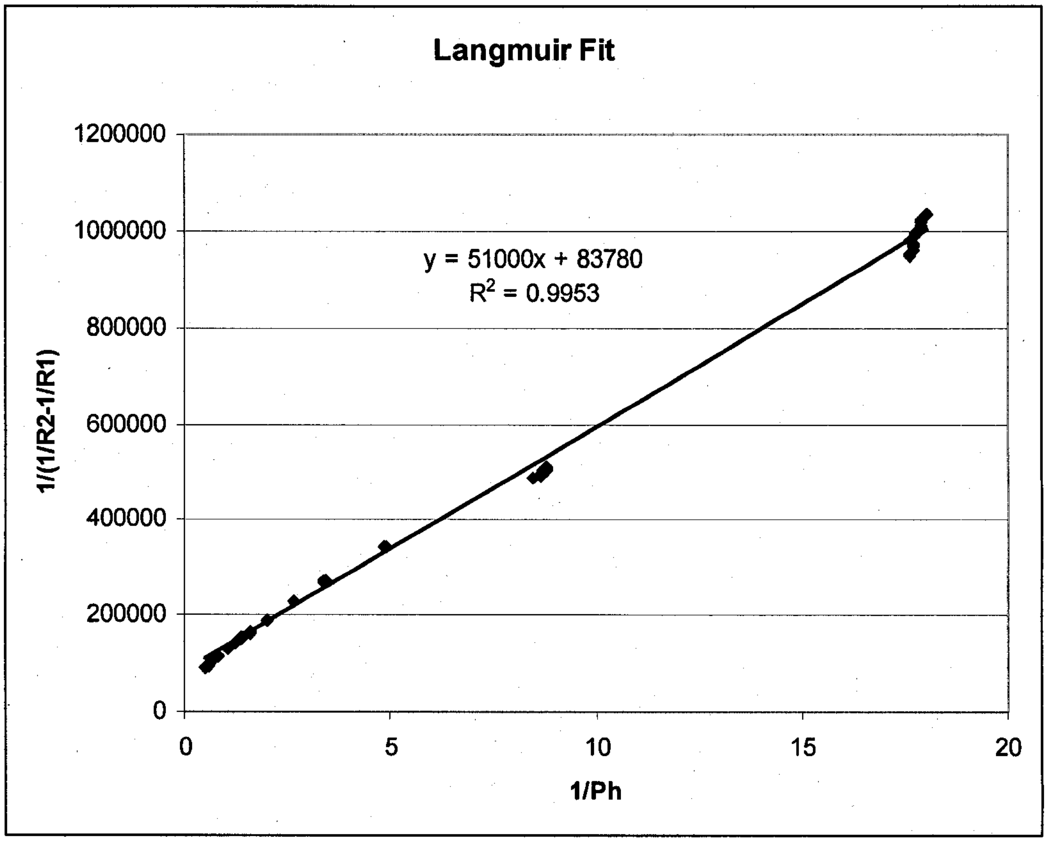


Figure 18: Langmuir fit for humidity at zero ozone concentration

The saturation resistance is then 35215 ohms, and the b value is 0.6087 kPa. The fit above suggests that the humidity response also fits a Langmuir isotherm, which will be used in the steady state analysis for both gases. However, the transient response to humidity does not follow a Langmuir fit: when the humidity increases or decreases there is an overshoot before the resistance comes back to its steady state value. This overshoot seems to be proportional to the change in humidity; for small changes (less than 2%RH), the overshoot is negligible, while for large changes (greater than 10%) the overshoot can be upwards of 5000 ohms. The humidity data was therefore not fit to the transient Langmuir equation. It is unclear why the sensor responds this way.

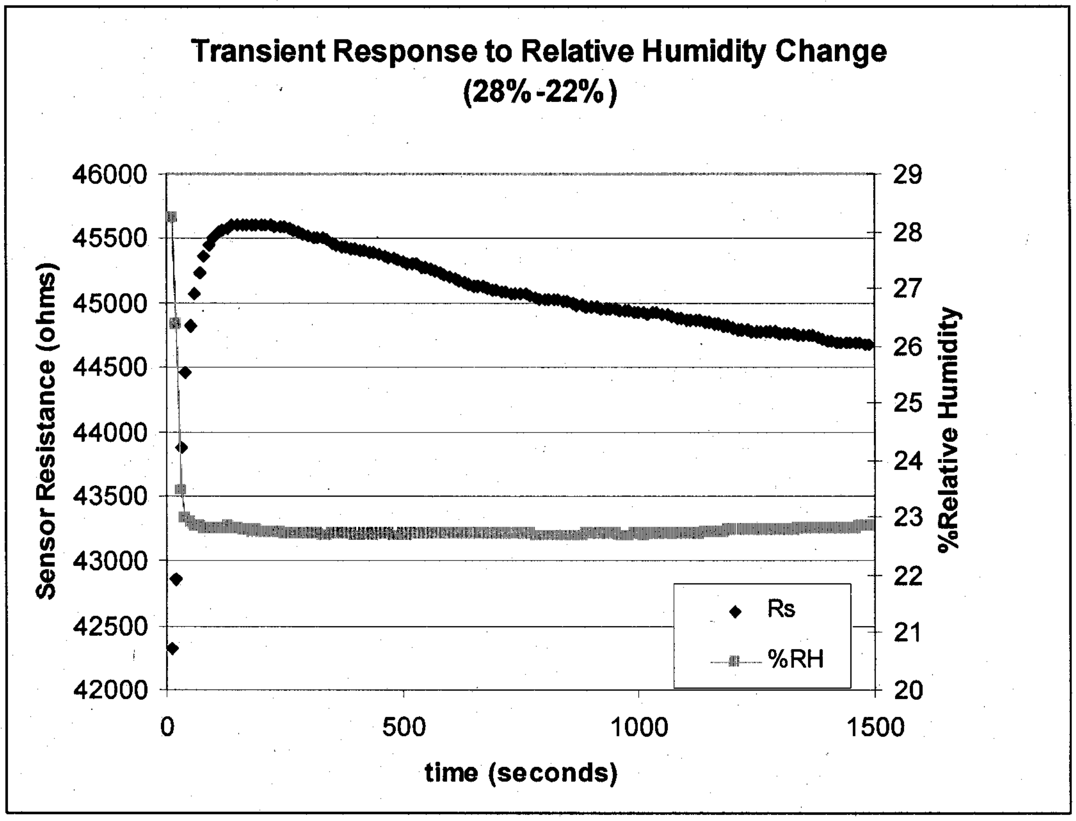


Figure 19: Sensor Response due to a change in humidity.

Langmuir Fit of Combined Data

The combined humidity and ozone data (figure 14) were fit to the adsorption equation using a MATLAB program ***lang.m*** (see Appendix A). The program finds the best fit to the data of the equation

$$R_2 = \frac{1}{-A\left(\dfrac{N_{t,oz}\dfrac{P_{oz}}{b_{oz}} - N_{t,w}\dfrac{P_w}{b_w}}{1+\dfrac{P_{oz}}{b_{oz}}+\dfrac{P_w}{b_w}}\right)+\dfrac{1}{R_1}} \tag{33}$$

by using nlinfit, a MATLAB function that varies the parameters (starting at an initial guess) to minimize the residuals between the fitted curve and the input data. Note that this is the equation assuming a different N_t for water and ozone. The program was originally run with equation 32 (the exact Langmuir isotherm equation), but a solution could not be found. For the current MATLAB program, The term $AN_{t,w}$ was combined into one constant, A, and the constant in front of the ozone term was redefined as C, so the fitted curve $f(P_{oz}, P_w)$ in the MATLAB program was

$$f(P_{oz}, P_w) = \frac{1}{A\left(\dfrac{C\dfrac{P_{oz}}{b_{oz}} + \dfrac{P_w}{b_w}}{1+\dfrac{P_{oz}}{b_{oz}}+\dfrac{P_w}{b_w}}\right)+\dfrac{1}{R_1}} \tag{37}$$

The relative humidities and the ozone concentrations were converted to partial pressures (kPa) for the calculation, using an average chamber temperature of 25°C and an average chamber pressure of 101.3 kPa. Also, as there were over 10,000 data points (the program can't accommodate that many), the data was pared down to 185 points. This was done first by only using the last minute of data at each RH/ozone combination (down to 2100 points) to ensure steady state had been achieved, and then by random selection. The results of the program are plotted in Figure 20. The circles are the data points; the crosses are the fitted curve points (each one corresponds to and should lie inside a blue circle).

The calculated parameters are

$$A = 3.7565e\text{-}5 \text{ /ohms}$$
$$b_{oz} = 1.77177e\text{-}6 \text{ kPa}$$
$$C = -0.406845$$
$$b_w = 4.7652 \text{ kPa}$$
$$R_l = 62392.97 \text{ ohms}$$

The R^2 value for the data was 0.9963, and the standard deviation was 3920 ohms. See MATLAB program for calculations.

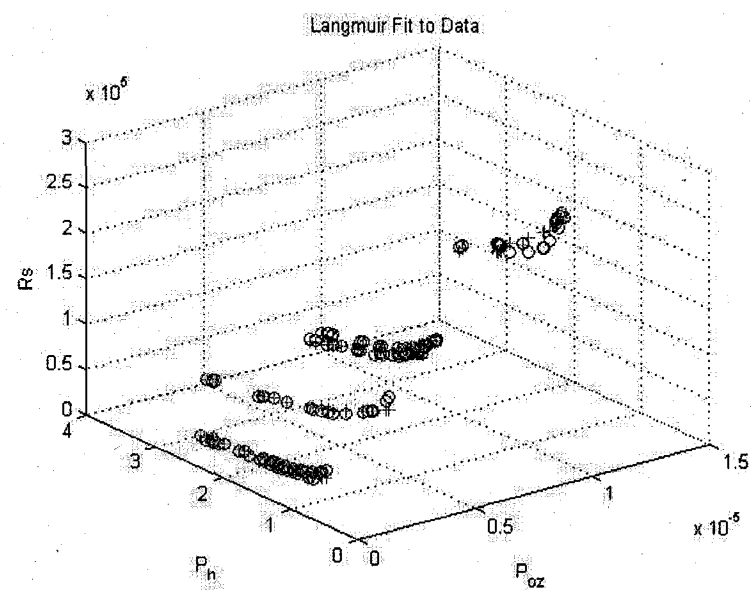


Figure 20: 3-D plot of curve fit (crosses) to data (circles).

Note that the b value calculated from the combined data (1.772e-6 kPa) is of the same order of magnitude of the two b values calculated from the low-humidity ozone data (1.642e-6 kPa and 2.33623e-6 kPa). The humidity data did not correlate quite as nicely: the b value calculated from the humidity data alone was 0.6087, while the combined data gave a b value of 4.7625 (difference of 682%). The difference is not due to using the full humidity range (note that in the humidity section, RH-15 was used instead of RH), as the b values should be the same despite using a different starting value. (To check, the program was re-run using RH-10%, and a b_w value of 6.01 was obtained- a difference of 20.7%.) It is unclear why the fits do not match.

The absolute value of C can be interpreted as the ratio between the number of adsorption sites of ozone to the number of adsorption sites of water, or the ratio of donated electrons (see section entitled *Equation Relating Partial Pressure to Sensor Resistance*). It was noted that the C value of -0.41 is close to -0.5, which could mean that water has exactly twice the number of adsorption sites as ozone, or that water donates twice the number of electrons.

The MATLAB program ***lang.m*** was re-run to check the fit with C specified to be -0.5. More data points were used (the program was adjusted to accommodate the 2100 data points), and with this parameter fixed an R^2 value of 0.9969 and a standard deviation of 4198ohms was achieved. Because the R^2 value was higher that the previous one, the program was re-run again using the same data points but without specifying C. This way achieved an R^2 value of 0.9970, and a standard deviation of 4151 ohms.

	Original	**Fixed C = -0.5**	**Recalculated**
A (ohms)	3.7565e-5	3.1209e-5	4.5210e-5
b_{oz} (kPa)	1.7718e-6	1.5944e-6	1.7773e-6
C	-0.4068	-0.500	-0.3285
b_w (kPa)	4.7652	3.6377	5.3515
R_l (ohms)	62393	60799	63989
R^2	0.9963	0.9969	0.9970
Std Dev	3920	4198	4151

Table 1: Calculated parameters for equation (37).

Note that defining a C value causes the parameters to have minor adjustments, but all stayed within the same order of magnitude. Because altering the C value to -0.5

created an equivalent calibration, it is thought that this estimate for C could be a good assumption. As stated previously, the absolute value of C could be the ratio of the number of donated electrons. This ratio would mean that water donates two electrons per adsorbed molecule while ozone takes away one. Because none of the adsorption models predict that water will donate two electrons, this would indicate that a new adsorption model needs to be developed, perhaps one which also includes an explanation of the sensor's transient response to water.

If the ratio is due to the number of adsorption sites, it could be related to a simple size difference in the adsorbed molecules. A single-bond radius of oxygen is 65.9pm, while hydrogen's is 29.9pm [8]. The sites may be close enough that an adsorbed ozone will block adjacent sites that aren't blocked when a hydrogen atom adsorbs.

Calibration Equation

To create a calibration equation for the sensor, equation 33 was rearranged to isolate Poz. The ozone concentration in parts per billion (x) was then substituted for the partial pressure of ozone so that the calibration would read out in the units of ppb.

$$x = \left(\frac{-b_{oz} \cdot 10^9}{P_{atm}}\right)\frac{R_1 b_w - R_1 P_w + AR_2 R_1 P_w + R_2 b_w + R_2 P_w}{b_w (R_2 + ACR_2 R_1 - R_1)} \qquad \textbf{(38)}$$

At 85%RH and 100 ppb, the standard deviation of 3920 ohms gives an error of approximately 4ppb, or 4% (lower humidities and lower ozone levels yield less absolute error – the 3920 ohm deviation correlates to an error of 1.7ppb at 20%RH and 10ppb).

The LabVIEW program was modified to record the ozone concentration calculated from the calibration equation, and a new set of tests were run to check its accuracy. The 50ppb tests were run first, then 25 ppb, then 100ppb, and finally 0ppb. The ozone values obtained from the sensor calibration were compared to the values obtained from the ozone analyzer. The following table shows the results of that experiment. The analyzer reading was subtracted from the sensor reading to get the difference in ppb. The percent error of the reading was calculated from the absolute value of this difference divided by the analyzer reading.

%RH	Ozone (ppb) Analyzer	Ozone (ppb) Sensor	Difference (ppb)	%error of reading
18.8	0.1	-0.9	-1	N/A
30.1	0.4	-1.7	-2.1	N/A
45.9	0.5	-2.3	-2.8	N/A
76.5	0.6	-1.2	-1.8	N/A
24.8	23.9	23.5	-0.4	1.67
32.3	23.8	23	-0.8	3.36
50	23.8	23	-0.8	3.36
66.1	25.1	24.3	-0.8	3.19
26	51.8	51.6	-0.2	0.39
70	53.3	56	2.7	5.07
24.7	102.7	104	1.3	1.27
54.5	107	109	2	1.87
70.9	108	118	10	9.26

Table 2: Calibration results

Note that the percent errors for the zero ozone values are not valid (the calculation would require dividing by zero); for low ozone values a $\pm$ ppb value should be used instead. The data shows that with the exception of the data point at 108 ppb and 70.9%RH, the fit is good to within $\pm$ 2.8ppb or 5% of the reading.

The graph below shows the results plotted as the predicted ozone concentration against the actual ozone concentration. If the calibration were exact, the data would all fall on the line y =x. The ozone analyzer has an error of $\pm$ 1 ppb, which is shown in the x-error bars. The y-error bars show the 5% error of the reading needed to obtain EPA standards.

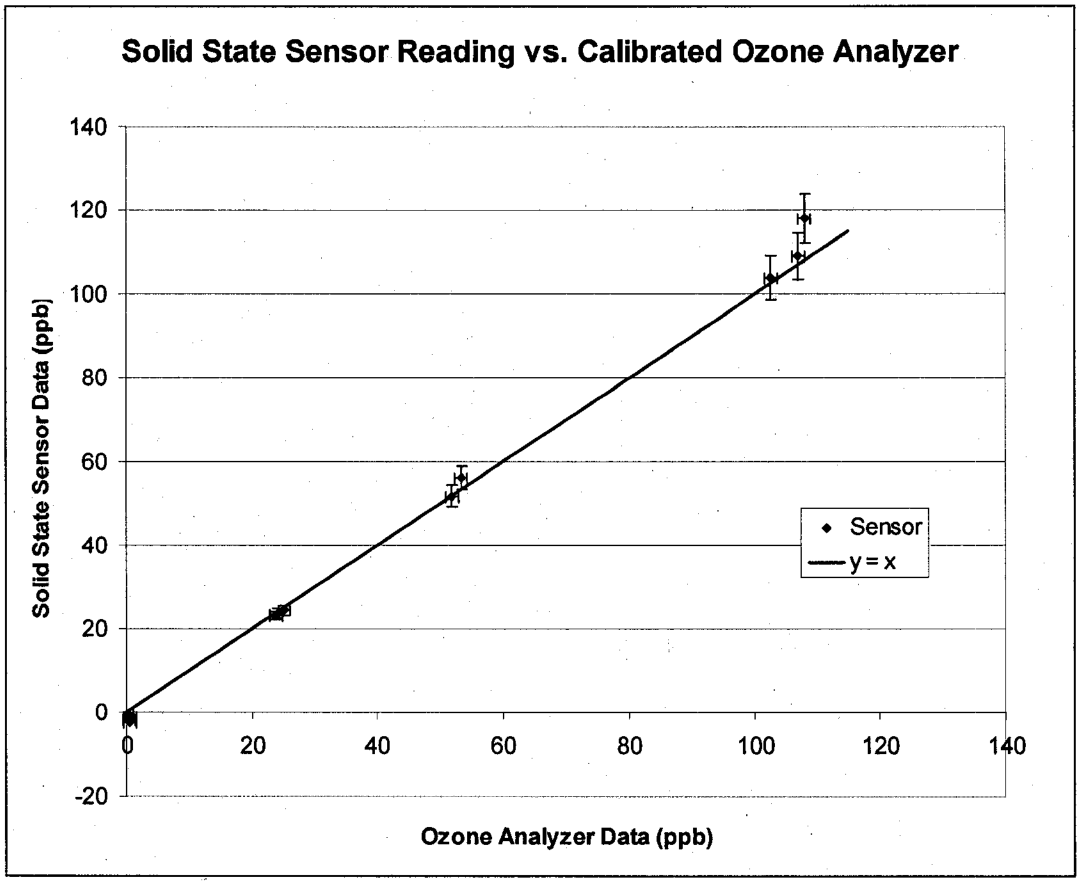


Figure 21: Sensor reading vs Actual reading. x-error bars indicate the precision of the analyzer measurement, while y-error bars indicate 5% error of the measurement.

CHAPTER VI

CONCLUSION

The acquired steady-state ozone data fit a Langmuir isotherm with an R^2 value of 0.9958. This suggested the use of a Langmuir equation to fit the data. Using equation (33) with a zero humidity assumption, the b_{oz} value was 1.642e-6 kPa, and the calculated saturation resistance was 1187252 ohms. Equation (20) was used to fit the transient data to the Langmuir model, yielding the rate coefficients α and β (2939.6 $(kPa\text{-}s)^{-1}$ and 0.00694 s^{-1}, respectively). If there was a way to accurately determine the sensor temperature, the physical constants that comprise α and β could be determined.

The acquired steady-state humidity data fit a Langmuir isotherm with an R^2 value of 0.9953, while the transient data suggests that a Langmuir model is inappropriate. Again, as we are interested in the steady state condition, the Langmuir model was used. The calculated b_w value was 0.6087 kPa and the saturation resistance was 35215 ohms.

The combined humidity and ozone data were fit to equation (37) using a MATLAB program. The The fit results were

$$A = 3.7565e\text{-}5 \text{ /ohms}$$
$$b_{oz} = 1.77177e\text{-}6 \text{ kPa}$$
$$C = -.406845$$
$$b_w = 4.7652 \text{ kPa}$$
$$R_l = 62392.97 \text{ ohms}$$

Note that the b_{oz} value was in between the two b_{oz} values calculated from low humidity data, while the calculated b_w constant was an order of magnitude higher than the one calculated from the zero-ozone humidity data. It is unclear why this occurred.

The combined humidity and ozone equation fit from MATLAB was rearranged to obtain an equation for in-situ calculation of the ozone concentration. This was input into the LabVIEW program and tested against the ozone analyzer. With the exception of one extraneous data point, the calibration held to within ± 2.8ppb, or ±5% of the reading.

CHAPTER VII

FUTURE WORK

Field Testing

The sensor will be set up with the pump and recalibrated for field testing. For this new calibration, the power will be adjusted to 82.5mW instead of 80mW. This will be done to compensate for the cooling effect of the air flow. (The manufacturer suggests 80mW in still air to achieve the recommended 430°C; the sensor resistance in still air was measured, then the pump turned on and the power varied until the sensor resistance matched the zero flow value.) This should reduce the time constant of the sensor, which will be good for stations that experience rapid changes in ozone concentration. The pump flow rate was tested at 0.54 ± 0.01L/min.

Extended humidity testing

The current setup has difficulty producing humidities over 70-75%RH, which means that the range between 75% and 100% may not have an accurate calibration. It was thought that the calibration would hold in this range despite the lack of test data, but in future calibrations this should be remedied if the field testing reveals that the calibration is inaccurate at high humidities.

Temperature Testing

The sensor will be used at weather stations and in weather balloons, where the ambient temperature will not remain constant. The sensor's response in different ambient temperatures needs to be tested. From the 1-D heat transfer equation

$$q_{gen} = \frac{h}{t}(T_{heater} - T_{ambient}) \quad \textbf{(39)}$$

where q_{gen} is the power generated per unit volume of the heater, h is the heat transfer coefficient, and t is the thickness of the heater, it can be assumed that the heater temperature will rise and fall in sync with the ambient temperature.

Because the sensor responds differently at different temperatures (see figure 4), there may need to be multiple calibration equations for different temperature ranges. Another option would be to adjust the power to compensate for the temperature change, requiring a power vs ambient temperature calibration.

Pressure Testing

The sensor may eventually be used in measurements taken in weather balloons that fly at high altitudes, and subsequently lower atmospheric pressures. To ensure that the sensor will accurately detect ozone levels at those high altitudes, pressure testing should be done. Also, oxygen concentrations may differ at those high altitudes, which could affect the sensor response (recall that oxygen is assumed to saturate the surface at normal atmospheric pressure).

Fitting Data to Other Isotherms

The data from the experiments was only fitted to the Langmuir isotherm. It happened to be the first one tested, and fit the steady-state data well enough to achieve a calibration curve that predicted ozone concentration to within 3ppb. However, the transient data, especially for humidity, did not fit a Langmuir model. This suggests that another adsorption model might be more suited for use with the data. Other isotherms, as well as polynomial and power equations should be tried to achieve a more accurate model of the adsorption process.

APPENDICES

APPENDIX A: MATLAB code

MATLAB Program temp_cycle.m

```
clear all;
close all;
clc;

theta = 0; %fraction of surface coverage
P = 5e-3; %Partial pressure (Pa)~50ppb
T = 273+410; %Temperature (K)
b = 0; %modified with a_and_b function
a = 0; %modified with a_and_b function
t= 0;
dt = .1;

tm = (0:dt:1000);
theta_new = zeros(1,10001);
theta_matrix = zeros(10,10001);
count2 = 1;

while (T < 273+450)
    count = 1;
    theta = 0;
    while ( count < 10002)

        [a,b,alpha,beta] = a_and_b (T, P, theta);
        d_theta = (alpha*P*(1-theta)-beta*theta)*dt;
        theta_new(count) = theta + d_theta;
        theta = theta_new(count);
        count = count +1;

    end
    theta_matrix(count2,:)=theta_new;
    plot(tm,theta_new)
    xlabel('time (seconds)')
    ylabel('fractional surface coverage')
    hold all
    count2 = count2+1;
    T = T+10;
end
```

The program uses the rate equation (equation 19) incremented in steps to create values of theta at different times. It uses a_and_b.m to calculate the a and b values for the rate equation. It saves the d_theta values in a matrix (theta_matrix), that has one row for each d_theta value. It then steps through the temperatures using another while loop, creating a new column for each temperature. At the end it plots each row of the theta_matrix against tm, the time vector, generating the graph of the isotherm at various temperatures.

MATLAB Program a_and_b.m

```
function [a, b, alpha, beta] = a_and_b (T, P, theta)

Kb = 1.38e-23; % boltzmanns constant (m^2kg/(s^2K))
m = 48/(6.02e23*1000); % mass of molecule (kgrams) =48
Ed = 2.25*1.602e-19; % energy of desorption (J/molec) 100KJ/mol=1.66e-
19J/molec - should be ~2
nu = 1e14; % rate of desorption (s^-1) should be ~1e14 (3.298e13for
oxygen on SnO2,1.4e14 for oxygen)
K = .01; %initial sticking probability 0-1
s = 1e-20; %fraction of molecules with enough energy to adsorb 0-1
Ks = K*s;%*(1-theta)^2; %sticking probability

alpha = Ks/(2*pi*m*Kb*T)^.5;
beta = nu*exp(-Ed/(Kb*T));

a = alpha*P + beta;

b = beta/alpha;
```

This program calculates the a and b values (and alpha and beta) given the temperature and pressure.

MATLAB Program fitlang.m

```
function [estimates, model] = fitlang()

clear all;
close all;
clc;

%Get data
channel = ddeinit('excel', 'oz_ORH.xls');
time = ddereq(channel, 'r107c11:r201c11');
saturation = ddereq(channel, 'r107c12:r201c12');
start_point = [.01];%start point for fminsearch
model = @ozone;
options = optimset('MaxIter',10000,'MaxFunEval',10000);
estimates = fminsearch(model, start_point,options);
% ozone accepts curve parameters as inputs, and outputs sse,
% the sum of squares error for the function,
% and the FittedCurve. FMINSEARCH only needs sse, but we want to
% plot the FittedCurve at the end.
    function [sse, FittedCurve] = ozone(params)
        P = 4.08e-6;%40.5ppb 4.08e-6 kPa
        b =2.36228e-6;%params(1);
        %alpha = params(1);%Ko*s/(2*pi*m*Kb*T)^.5;
        bet = params(1);%nu*exp(-Ed/(Kb*T));
        %bet =b*alpha;
        alpha = bet/b;
        a = alpha*P + bet;
        %b = bet/alpha;
        FittedCurve = 1./(1+b/P)*(1-exp(-a*time));
        ErrorVector = FittedCurve - saturation;
        sse = sum(ErrorVector .^ 2);
    end

[sse, FittedCurve] = model(estimates);

%plot data and fitted curve
hold all
plot(time, FittedCurve)
plot(time,saturation,'+')
xlabel('time')
ylabel('theta')
end
```

This program gets data from an excel file, and uses a defined function **ozone** to calculate the sum of squared errors between the fitted curve (defined in the function) and the excel data. Ozone accepts an input parameter for one variable and returns the sum of squared errors (and the fitted curve). The ozone function is input into fminsearch, a MATLAB function that minimizes the input function's output (in this case sse) by changing the variables (in this case params). It will output the variable that created the smallest sse. This variable is then input into **ozone** again to generate a plot using the fittedcurve, and the data is also plotted to visualize the goodness of fit.

MATLAB Program fit_ab.m

```
%clear all;
%close all;
%clc;

theta = .63316; %fraction of surface coverage
P = 8.3e-6; %Partial pressure (kPa)~80ppb

b = 2.36228e-6;%calculated from data
%alpha = 2939.56; %K/(2*pi*m*Kb*T)^.5;
beta = .0069; %nu*exp(-Ed/(Kb*T));.0047455

alpha = beta/b;
%beta = b*alpha;

t= 0;
dt = .01;
tm = (0:dt:750);
theta_new = zeros(1,75001);
theta_matrix = zeros(10,75001);
count2 = 1;

count = 1;
while ( count < 75002)
   d_theta = (alpha*P*(1-theta)-beta*theta)*dt;
   theta_new(count) = theta + d_theta;
   theta = theta_new(count);
   count = count +1;

end
theta_matrix(count2,:)=theta_new;
plot(tm,theta_new)
xlabel('time (seconds)')
ylabel('fractional surface coverage')
hold all

channel = ddeinit('excel', 'oz_0RH.xls');
time = ddereq(channel, 'r2c11:r100c11');
saturation = ddereq(channel, 'r2c12:r100c12');
plot(time,saturation,'+')
```

This program plots data vs. a curve generated from previously defined constants.

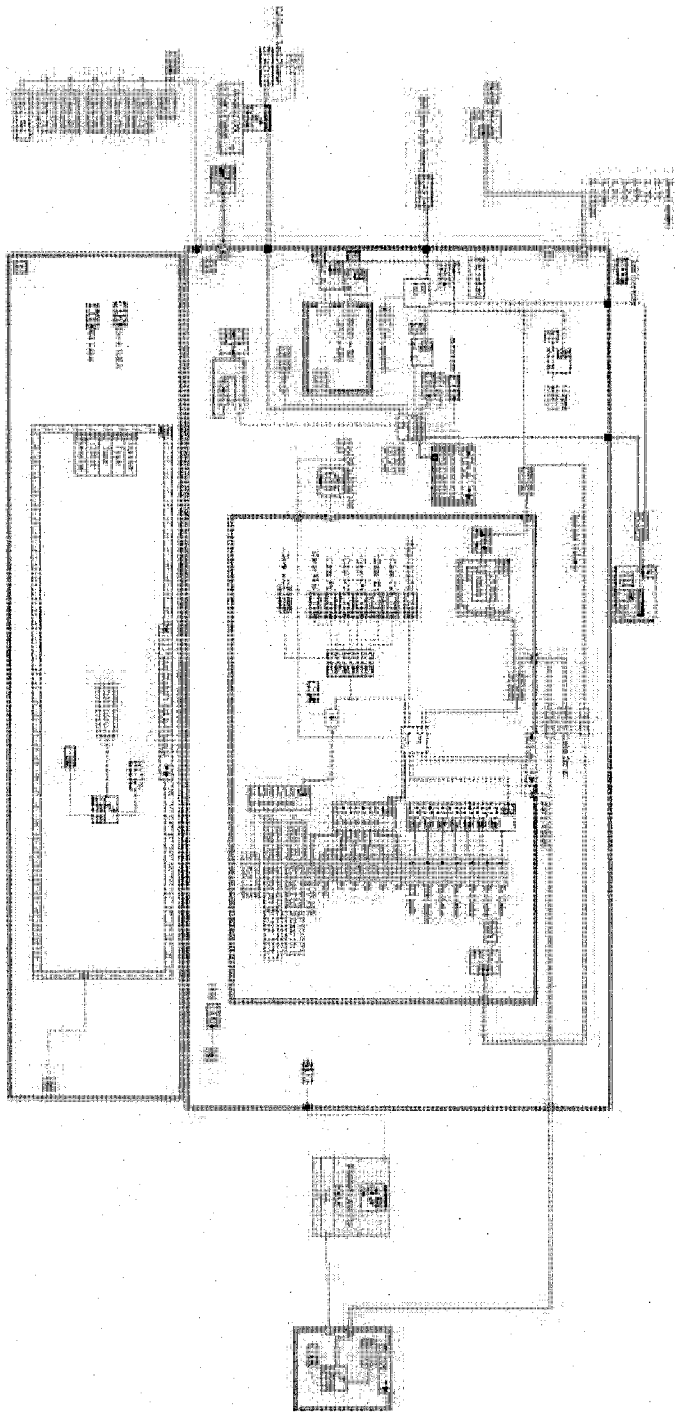
MATLAB Program lang.m

```
function [paramfit, Rsquare] = lang()

clear all;
close all;
clc;

%Get data
channel = ddeinit('excel', 'RH_oz.xls');
resistance = ddereq(channel, 'r1c14:r185c14');
oz = ddereq(channel, 'r1c13:r185c13');
RH = ddereq(channel,'r1c15:r185c15');
T = 25;%ddereq(channel,'r2c1:r260c1');
P = 760;%ddereq(channel,'r2c5:r260c5');

%Convert oz and RH to pressure kPa
Poz = oz.*10^-9.*P.*0.1333;
Ph = RH./1000.*(6.1078+T.*(0.44365+T.*(0.014299+T.*(0.00026506+
T.*(3.03124*10^-6+T.*(2.034*10^-8+T.*6.1368*10^-11))))));

%change form of variables so they can be used with nlinfit, and provide
%initial guess
variables = [Poz, Ph];
beta = [.0000203,.00000139,-.5,3.6,64500];

%define equation
    function ozone = langmuir(beta,variables)
       A = beta(1);
       Boz = beta(2);
       C = beta(3);
       bH = beta(4);
       Ro =beta(5);

       ozone = 1./(A*(variables(:,2)./bH + C*variables(:,1)./Boz)./(1 +
variables(:,2)./bH + variables(:,1)./Boz)+ 1./Ro);
    end

%use nlinfit to find best fit for constants in equation - have to use
%options to increase the number of iterations so program doesn't choke.
%nlinfit outputs paramfit, which is a vector containing the best fit
%constant values, and r, which is a vector containing the error at each
%point.
options = statset('MaxIter',1000);
[paramfit,r] = nlinfit(variables, resistance,@langmuir,beta,options);

%find R^2 value
i = 1;
sse=0;
while i<length(resistance)
    sse=sse+r(i)*r(i);
    i=i+1;
end
```

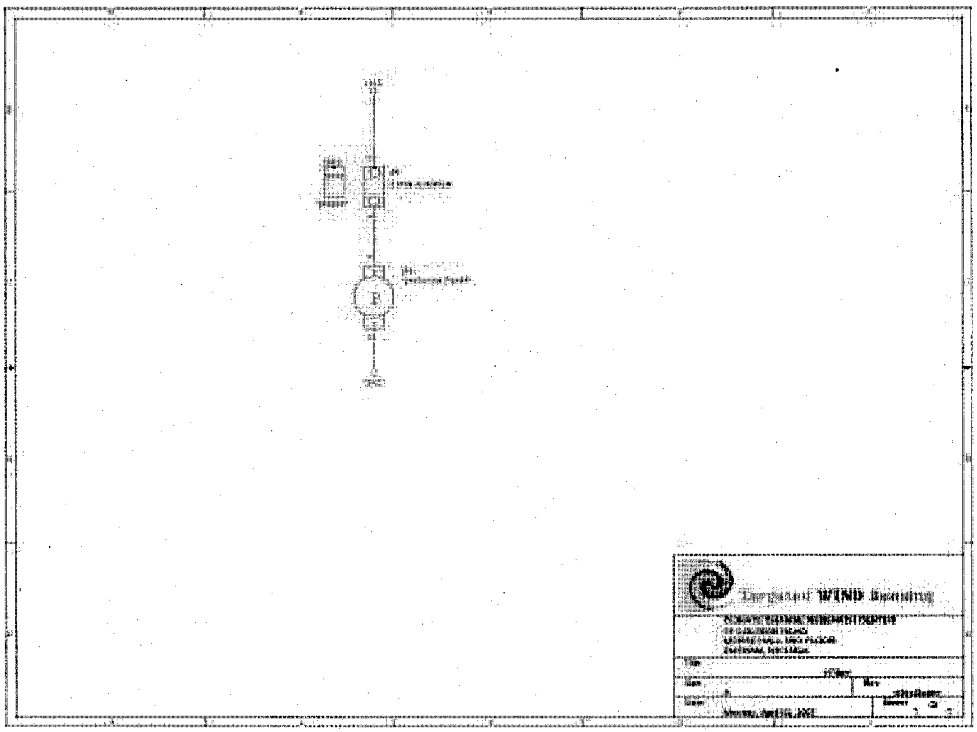
```
M = mean(resistance);
diff = (resistance - M).^2;
delta = sum(diff);
Rsquare = 1-sse/delta
DoF = length(resistance) - 5;
standard_dev = (sse/DoF)^.5

%plot data and fitted curve
scatter3(Poz,Ph,resistance)
hold on
fittedcurve = 1./(A*(Ph./bH + C*Poz./Boz)./(1 + Ph./bH + Poz./Boz)+
1./Ro);
scatter3(Poz,Ph, fittedcurve,'+')
title('Langmuir Fit to Data')
xlabel('P_o_z')
ylabel('P_h')
zlabel('Rs')
end
```

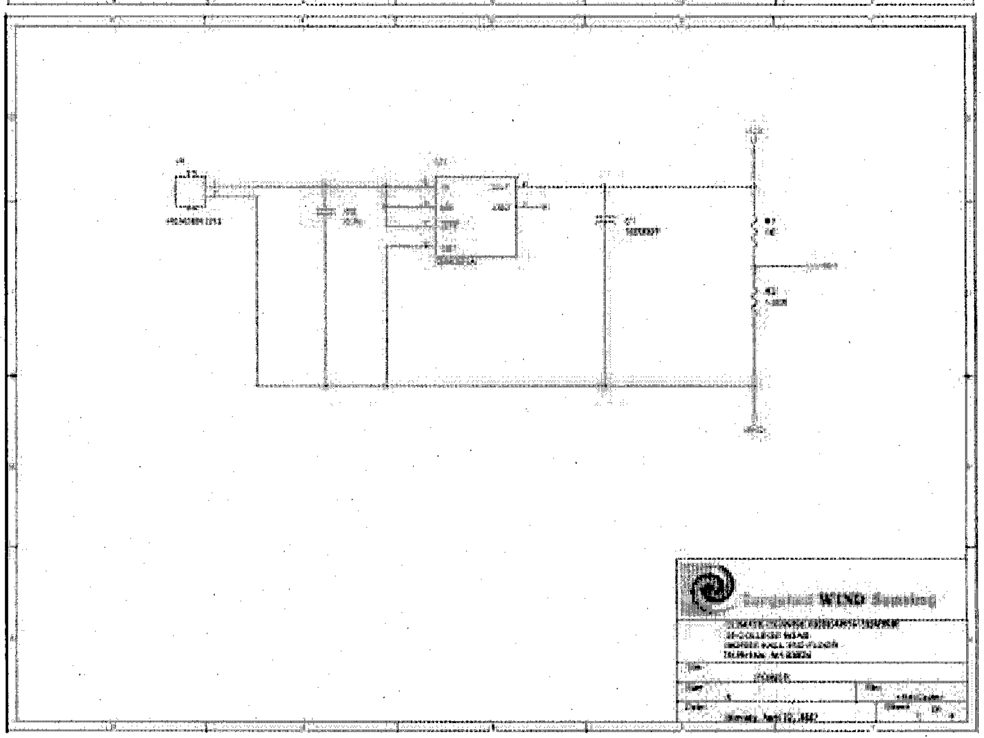
This program inputs data from an excel file and uses a defined function **langmuir** to calculate a predicted ozone value. **Langmuir**, the input data, and a set of initial guesses are input into the MATLAB function nlinfit, which finds the minimum of the residuals (the difference between the curve from **langmuir** and the data) by varying the parameters in the **langmuir** function. It then uses the residuals vector to find the R^2 value and standard deviation. (See code for the equations). After that, it plots the data and the curve fit onto a 3-d plot.

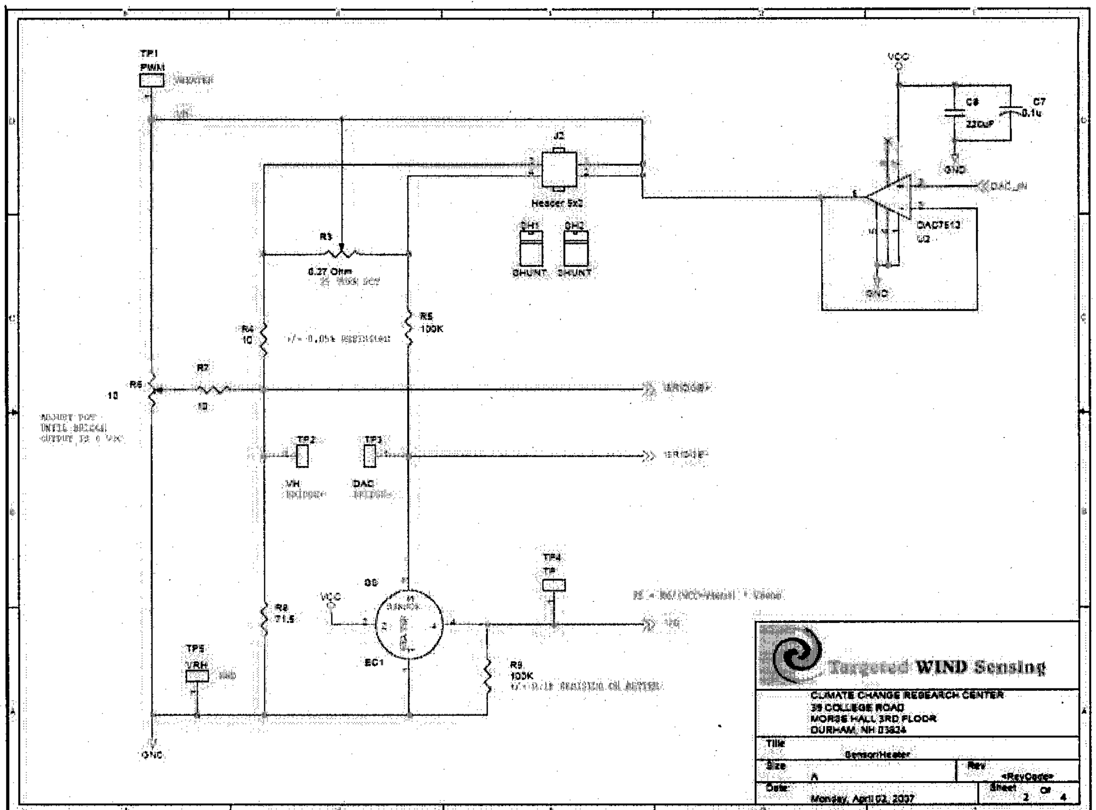
APPENDIX B: LabVIEW Program

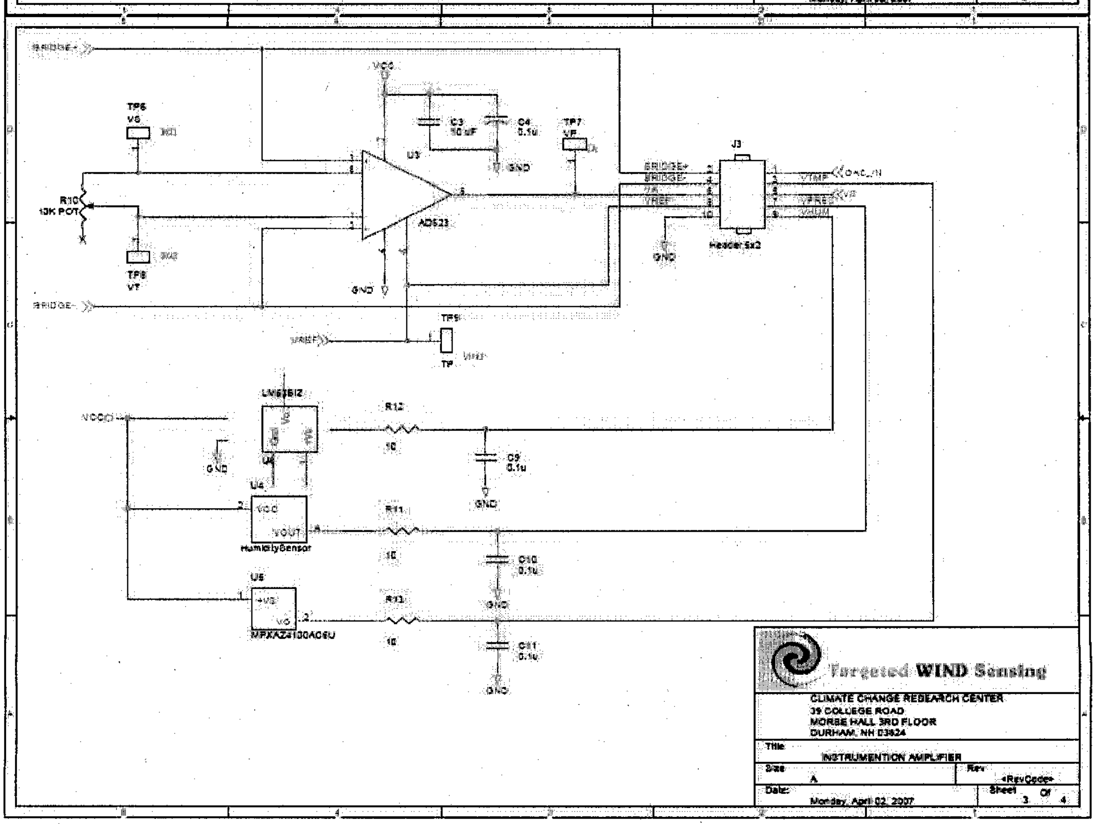


APPENDIX C: Circuit Diagram






TP1
PWM
VCC
C8
220uF
C7
0.1u
GND
J2
Header 5x2
DAC_IN
DAC7513
U2
SH1
SH2
SHUNT
SHUNT
R3
0.27 Ohm
R4
10
R5
100K
R6
10
R7
10
BRIDGE+
BRIDGE-
TP2
VH
TP3
DAC
TP4
TP
VCC
R8
71.5
R9
100K
TP5
VRH
EC1
GND
Targeted WIND Sensing
CLIMATE CHANGE RESEARCH CENTER
39 COLLEGE ROAD
MORSE HALL 3RD FLOOR
DURHAM, NH 03824
Title
Sensor/Heater
Size
A
Rev
<RevCode>
Date:
Monday, April 02, 2007
Sheet 2 of 4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TP6
VG
VCC
C3
10 uF
C4
0.1u
TP7
VF
GND
J3
R10
10K POT
U3
AD623
BRIDGE+
BRIDGE-
VF
VREF
VTMP
VPRES
VHUM
DAC_IN
Header 5x2
TP8
VT
GND
BRIDGE-
TP9
VREF
TP
LM62BIZ
VCC
GND
U4
R12
10
C9
0.1u
GND
U4
VCC
VOUT
HumiditySensor
R11
10
C10
0.1u
U6
+VS
VO
MPXAZ4100AC6U
R13
10
C11
0.1u
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MORSE HALL 3RD FLOOR
DURHAM, NH 03824
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INSTRUMENTION AMPLIFIER
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Monday, April 02, 2007
Sheet 3 of 4

APPENDIX D: MiCS Ozone Sensor

MiCS

MicroChemical Systems

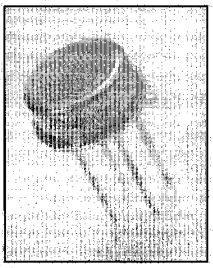
PRODUCT DATA SHEET

MiCS – 2610

O_3 Gas Sensor

This datasheet describes the use of the MiCS-2610 in ozone detection applications. The package and the mode of operation described in this document target the detection of the oxidizing gas O_3 in indoor or outdoor environments. Ozone is a hazardous gas, which can cause respiratory problems at concentrations above 100 ppb.

Features:



- Low heater current
- Wide detection range
- High sensitivity
- Fast thermal response
- Electro-Static Discharge protected
- Miniature dimensions
- High resistance to shocks and vibrations

For this and other quality MiCS products, send an e-mail to info@microchemical.com or contact MicroChemical Systems at:

Rue de Porcena 15 • CH-2035 Corcelles, Switzerland • Tel: 41 (0) 32 731 0120 • Fax: 41 (0) 32 731 01 24

Sensor Characteristics

Important Precautions:

Please read the following instructions carefully before using the MiCS-2610 sensor described in this document to avoid erroneous readings and to prevent the device from permanent damage.

- The sensor must not be wave soldered without protection or exposed to high concentrations of organic solvents, ammonia, or silicone vapors in order to avoid poisoning the sensitive layer.
- Heating powers above the specified maximum rating of 95 mW can destroy the sensor due to overheating.
- After exposing the sensor to high concentrations of O_3, make sure the sensor is given enough time to recover before taking new measurements.
- For any additional questions, please contact us at: apps@microchemical.com

Operating Mode:

The recommended mode of operation is a constant voltage mode. A heating power of P_H = 80 mW is applied. This causes the temperature of the sensing resistor (R_S) to reach about 430°C.

Detection of the O_3 concentration is achieved by measuring the sensing resistor R_S during operation.

Sensor Response:

The sensor response to O_3 in air is represented in Figure 1. The sensor resistance R_S is normalized to the resistance under 100 ppb of O_3 (R_{100ppb}).

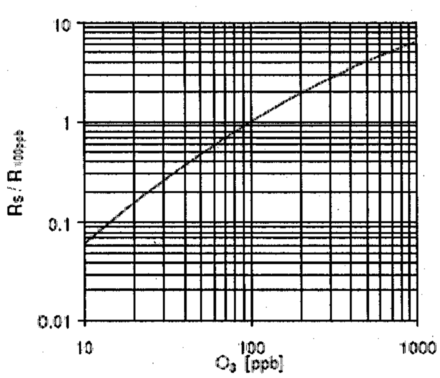
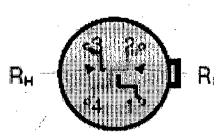


Figure 1: R_S / R_{100ppb} as a function of gas concentration at 50% RH and 25°C.

Measurement Circuit:

Figure 2 shows the pin connections of the MiCS-2610 ozone sensor. A simple circuit to measure the O_3 concentration is proposed in Figure 3. The heating voltage V_H is applied to pins 3 and 1. A load resistor R_L is connected in series with R_S to convert the resistance R_S to a voltage V_S between pins 2 and 4. R_S can then be calculated by the following expression:

$$R_S = R_L / (V_{CC} - V_S) \cdot V_S$$



Pin Number	
1	Heater Ground
2	Sensor Pin
3	Heater Power
4	Sensor Pin

Figure 2: Equivalent circuit (top view) of MiCS 2610.

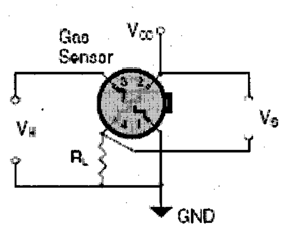


Figure 3: Measurement circuit for O_3 detection.

Electrical Specifications

Maximum Ratings:

Rating	Symbol	Value / Range	Unit
Maximum Sensor Supply Voltage	V_{CC}	5	V
Maximum Heater Power Dissipation [1]	P_H	95	mW
Maximum Sensor Power Dissipation	P_S	1	mW
Relative Humidity Range	R_H	5 – 95	%RH
Ambient Operating Temperature	T_{amb}	-40 – 70	°C
Storage Temperature Range [2]	T_{sto}	-40 – 50	°C
Storage Humidity Range	RH_{sto}	5 – 95	%RH

Table 1

[1] Heating powers above 95 mW can cause permanent damage to the sensor due to overheating.

[2] Storage of parts in original shipping package

Operating Conditions:

Parameter	Symbol	Typ	Min	Max	Unit
Heating Power [3]	P_H	80	66	95	mW
Heating Voltage	V_H	2.35	-	-	V
Heating Current	I_H	34	-	-	mA
Heating Resistance [4]	R_H	68	58	78	Ω

Table 2

[3] To ensure a correct operating temperature the heater voltage should be ajusted so that the resulting heating power equals 80 mW. Lower heating power will reduce the sensitivity and increase the response time. Heating powers above 95 mW can cause permanent damage to the sensor due to overheating.

[4] Heating resistor values from sensors out of production range between 58 and 78 Ohm measured at V_H = 2.35 V. Due to material properties of the heating resistor its value increases during operating life.

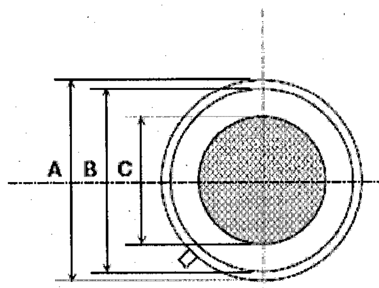
Sensitivity Characteristics:

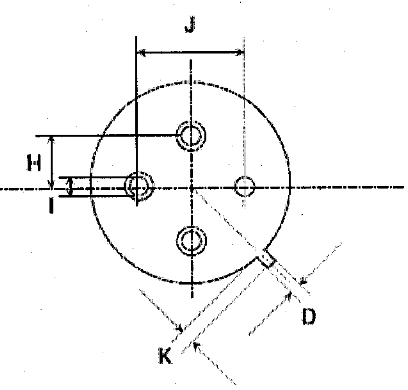
Characteristic	Symbol	Typ	Min	Max	Unit
O_3 Detection Range	FS		10	1000	ppb
Sensing Resistance in air	R_0	11	3	60	kΩ
Sensitivity Factor [5]	S_R	2	1.5	4	-

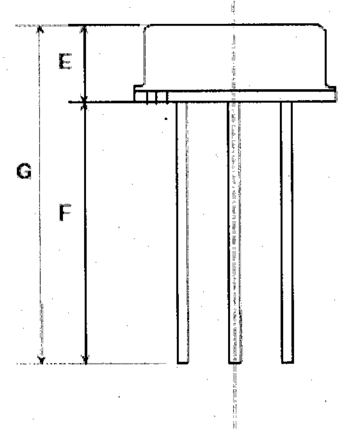
Table 3

[5] Sensitivity Factor S_R is defined as RS at 100 ppb of O_3 divided by RS at 50 ppb of O_3. Test conditions are 50±5 %RH and 25±2 °C.

Package Dimensions







Dimension	Min [mm]	Max [mm]
A	9	9.4
B	8.15	8.30
C	5.75	5.85
D	0.6	0.9
E	3.5	3.9
F	9.0	10.0
G	12.5	13.9
H	2.41	2.67
I	0.55	0.65
J	4.83	5.33
K	0.7	0.9

APPENDIX E: Flow Rate Calculation

$TubeLength := 3.20in$

$TubeRadius := 0.25in$

$FlowRate := 0.5\frac{L}{min}$

$TubeVolume := TubeRadius^{2} \cdot \pi \cdot TubeLength$

$TubeVolume = 0.01\,L$

$FlowRate = 8.333 \times 10^{-6}\,\frac{m^{3}}{s}$

$SlugFlowTime := \frac{TubeVolume}{FlowRate}$

$SlugFlowTime = 1.236\,s$

APPENDIX F: Calibration Data

Combined Humidity and Ozone data (RH_oz.xls)

ozone	resistance	RH
50.6	119182.2	45.66312
50.6	119617.6	44.7401
50.6	119645.2	44.75573
50.6	119746	44.76606
50.6	119840.7	44.74355
50.4	120569.4	44.12619
50.4	120576.3	44.07227
50.4	120717.2	44.0524
50.4	120795.9	44.02091
50.4	122643.6	42.44267
50.4	122517.2	42.42978
50.4	124008.4	41.2659
50.4	123951.8	41.28401
50.4	123936.1	40.98931
50.4	125301.1	39.99556
50.4	125440.6	39.8557
50.4	127332.6	38.24877
50.4	132270.8	34.26313
50.4	132023.8	34.1598
50.4	132045.1	34.15481
50.4	132881.7	33.28619
50.4	132949.7	33.26232
50	136342.7	30.82007
50	136754.7	30.79583
50	139160.3	29.26834
50	139297.6	29.21851
50	140019.6	28.63608
50	140346.3	28.46819
50	140723.1	28.42934
50	140882.6	28.29014
50	141632.8	27.7012
50	141857.6	27.61322
49.9	143418.7	26.94415
49.9	143756.1	26.73727
49.9	143958.2	26.71216
49.9	143661.7	26.73506
49.9	145095.2	25.86873
49.9	145168.9	25.85569
49.9	146969.4	24.9801
49.9	147155.5	24.86906
49.9	147479.3	24.71429
49.9	147138	25.18002
49.9	146756	25.36217
50.3	144598.3	26.22667
50.3	142120.5	27.33071
50.3	142034.9	27.32239
50.3	141655.6	27.3319
50.3	137643	29.78872
50.3	137356.5	29.92256
50.8	132546.4	32.79926
50.8	132433	32.80571
50.8	132346.9	32.80803
50.8	132082.8	32.84569
50.8	131990.6	32.85137
50.8	131960.1	32.85858
50.4	131980.3	32.90084
50.4	131489.2	33.37311
50.4	129722.9	35.24815
50.4	128628	35.74583
50.4	128343.4	35.74743
50.4	128126.4	35.75499
50.4	127978.8	35.93006
50.4	127770.4	36.18443
50.4	127650.7	36.22944
50.4	127619.1	36.24757
50.4	126067.1	37.51923
50.4	126081.5	37.52382
50.4	126052.1	37.52329
50.4	124529.1	38.96707
50.4	124424.1	39.08747
50.8	123474.4	39.86414
50.8	123320.9	40.09962
50.8	121321.9	42.22837
50.8	121276.8	42.25166
50.8	119324.6	44.30626
50.8	118348.1	45.37277
50.7	116131.6	47.93269
50.7	116122	47.95315
50.7	114755.5	50.11083
50.7	114696.8	50.11938
50.7	114698.1	50.12515
50.7	112914.8	52.86136
50.7	112846.2	52.8867
51	111807.8	55.36655
51	111699.1	55.3909
51	111603.3	55.42059
51	111580.6	55.42848
51	111593.1	55.42103
51.2	110533.5	57.00695
51.2	110561.8	56.99296
51.2	110599.1	56.98285
51.2	109798.6	58.59395
51.2	108959.8	60.24184
51.2	108585.6	60.62845
51.2	107624.4	62.04923
51.2	107595.1	62.04407
51.2	106509.6	63.82234
51.2	106513.9	63.8334
51.5	105910.9	64.57491
51.5	105863.8	64.57337
51.5	105872.1	64.5618
51	105519.8	65.88457
51	105164.5	66.59071
51	105057.3	66.67358
51	104205.8	67.82222
51	104213.6	67.84973
51	104215.9	67.85634
51.4	103057.1	69.05924
51.4	103280	69.05682
51.4	103158.9	69.05199
51.2	102332	70.08017
51.2	101848.5	71.23522
51.2	101692.4	71.79709
51.2	101709.4	71.94979
51.2	101013.3	76.10937
51.2	100859.6	76.2707
51.2	100709.8	76.3631
51.2	99765.61	78.09057
51.2	99320.45	79.14917
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0.1	32805.57	71.15517

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0.1	32813.28	71.13024
0.1	32881.73	71.02477
0.1	32884.76	71.0154
0.1	32882.21	71.01378
0.1	32995.67	70.78358
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0.1	33187.8	70.57938
0.1	33277.14	70.54153
0.1	33275.76	70.54301
0.1	33361.63	70.44645
0.1	33355.54	70.44844
0.1	33449.4	70.39452
0.1	33453.37	70.40528
0.1	33508.04	70.35781
0.1	33505.83	70.3432
0.1	33654.25	70.21849
0.1	33791.51	70.04506
0.1	33863.6	70.00079
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0.1	33900.29	69.87129
0.1	33914.95	69.82394
0.1	33917.94	69.7832
0.1	33911.21	69.82874
0.1	33910.21	69.83481
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0.1	34149.79	68.51723
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0.1	34156.7	68.48973
0.1	34201.63	68.29543

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0.1	34266.97	68.24326
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0.1	34342.35	68.02307
0.1	34367.74	68.05977
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0.1	34371.27	68.06754
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0.1	34505.08	67.79352
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0.1	34524.6	67.7679
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0.1	34530.23	67.70965
0.1	34564.59	67.60712
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0.1	34546.88	67.6805
0.1	34540.7	67.72004
0.1	34532.7	67.73775
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0.1	34569.25	67.59908
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0.1	34577.46	67.57989
0.1	34559.54	67.60493
0.1	34558	67.63565
0.1	34563.88	67.60914
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0.1	34654.63	67.32623
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0.1	34637.86	67.48573
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0.1	34530.52	67.3991
0.1	34539.33	67.40815
0.1	34663.24	67.4134
0.1	34651.78	67.49818
0.1	34643.64	67.47313
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0.1	34693.65	67.42256
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0.1	34715.36	67.29941
0.1	34718.49	67.29469
0.1	34720.39	67.25152
0.1	34737.54	67.26881
0.1	34745.94	67.24574
0.1	34743.48	67.31734
0.1	34743.71	67.32258
0.1	34746.97	67.25189
0.1	34731.87	67.3088
0.1	34744.32	67.31712
0.1	34745.16	67.35872
0.1	34747.26	67.3045
0.1	34740.45	67.33845
0.1	34737.78	67.3406

0.1	34720.98	67.41974
0.1	34713.18	67.45816
0.1	34719.82	67.42557
0.1	34714.06	67.46244
0.1	34716.28	67.47445
0.1	34714.03	67.50312
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0.1	34683.64	67.59151
0.1	34694.55	67.51853
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0.1	34688.2	67.57829
0.1	34681.76	67.62135
0.1	34691.04	67.52845
0.1	34668.63	67.58563
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0.1	34700.66	67.5836
0.1	34676.76	67.6442
0.1	34670.24	67.60606
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0.1	34678.74	67.6535
0.1	34674.67	67.58174
0.1	34674.6	67.53671
0.1	34675.49	67.52186
0.1	34676.29	67.5227
0.1	34677.03	67.61663
0.1	34665.49	67.6357
0.1	34669.64	67.58039
0.1	34669.72	67.62277
0.1	34677.93	67.56541
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0.1	34676.74	67.55576
0.1	34671.61	67.58367
0.1	34683.54	67.4747
0.1	34698.09	67.40839
0.1	34705.38	67.39231
0.1	34705.32	67.46231
0.1	34695.44	67.48207
0.1	34697.7	67.40092
0.1	34692.65	67.49604
0.1	34696.63	67.49344
0.1	34697.27	67.4216
0.1	34700.22	67.4225
0.1	34718	67.47887
0.1	34710.86	67.53133
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0.1	34708.35	67.409

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0.1	34701.38	67.51833
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0.1	34698.26	67.46455
0.1	34697.7	67.49515
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0.1	34678.53	67.58928
0.1	34679.63	67.56768
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0.1	34688.76	67.52553
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0.1	34692.9	67.48625
0.1	34696.31	67.48614
0.1	34697.01	67.46869
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0.1	34707.93	67.4729
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0.1	34712.81	67.40745
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0.1	34717.43	67.48037
0.1	34710.2	67.50957
0.1	34712.71	67.44438
0.1	34715.46	67.4436
0.1	34718.41	67.45092
0.1	34711.52	67.47545
0.1	34712.28	67.50435
0.1	34691.52	67.54104
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0.1	34696.27	67.5128
0.1	34685.81	67.52231
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0.1	34704.45	67.4997
0.1	34703.94	67.52612
0.1	34703.55	67.47317
0.1	34706.05	67.48249
0.1	34709.69	67.4366
0.1	34716.93	67.4353
0.1	34716.24	67.46141
0.1	34707.16	67.47184
0.1	34706.16	67.43319

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0.1	34705.24	67.53564
0.1	34698.25	67.50151
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0.1	34709.75	67.36261
0.1	34708.79	67.48563
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0.1	34723.15	67.42124
0.1	34733.16	67.39194
0.1	34713.64	67.36924
0.1	34727.97	67.40216
0.1	34746.63	67.33489
0.1	34730.89	67.37199
0.1	34720.04	67.36168
0.1	34731.42	67.37274
0.1	34736.42	67.32651
0.1	34724.22	67.35321
0.1	34730.3	67.31083
0.1	34730.73	67.37131
0.1	34728.81	67.41246
0.1	34716.38	67.40779
0.1	34727.93	67.47095
0.1	34714.05	67.46675
0.1	34710.62	67.48187
0.1	34707.49	67.49607
0.1	34711.75	67.48587
0.1	34699.28	67.53505
0.1	34692.55	67.47196
0.1	34713.24	67.48452
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0.1	34705.84	67.49162
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0.1	34710.73	67.43285
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0.1	34717.13	67.36069
0.1	34730.02	67.38825

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0.1	34713.77	67.30052
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0.1	34737.23	67.39319
0.1	34732.07	67.35855
0.1	34735.66	67.42041
0.1	34714.61	67.54812
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0.1	34700.85	67.48125
0.1	34699.35	67.49925
0.1	34692.68	67.56092
0.1	34693.75	67.54306
0.1	34698.34	67.43525
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0.1	34710.78	67.41254
0.1	34708.4	67.50581
0.1	34707.21	67.45076
0.1	34713.65	67.52152
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0.1	34719.58	67.44678
0.1	34701.9	67.44357
0.1	34707.11	67.54784
0.1	34690.41	67.52588
0.1	34695.47	67.45864
0.1	34691.88	67.51893
0.1	34687.01	67.50972
0.1	34680.75	67.52593
0.1	34688.08	67.47796
0.1	34688.29	67.43349
0.1	34693.99	67.45931
0.1	34698.87	67.46379
0.1	34697.41	67.49596
0.1	34706.47	67.44576
0.1	34707.59	67.44092
0.1	34713.43	67.4238
0.1	34709.88	67.41715
0.1	34701.34	67.4061
0.1	34701.76	67.44682
0.1	34697.87	67.48556
0.1	34703.64	67.47129
0.1	34712.54	67.46973
0.1	34699.09	67.49156
0.1	34690.07	67.52733
0.1	34713.2	67.3869
0.1	34718.76	67.36593
0.1	34702.88	67.40205
0.1	34706.41	67.40063
0.1	34697.65	67.46345
0.1	34698.3	67.41271
0.1	34697.84	67.42405

0.1	34682.87	67.41883
0.1	34684.46	67.41878
0.1	34684.34	67.38157
0.1	34692.03	67.44142
0.1	34676.82	67.41683
0.1	34680.98	67.44774
0.1	34683.95	67.41825
0.1	34685.25	67.42241
0.1	34688.15	67.38321
0.1	34679.54	67.38089
0.1	34678.37	67.3771
0.1	34693.03	67.37324
0.1	34695.99	67.41751
0.1	34697.14	67.42227
0.1	34691.09	67.45327
0.1	34681.14	67.41457
0.1	34672.52	67.47241
0.1	34667.33	67.46708
0.1	34663.06	67.48698
0.1	34668.78	67.39289
0.1	34664.3	67.43676
0.1	34660.6	67.48784
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23.5	76644.53	48.78703
23.5	76645.73	48.79225
23.5	76610.48	48.87009
23.5	76594.8	48.86172
23.5	76604.03	48.85692
23.5	76608.02	48.88741
22.9	71775.24	58.3284
22.9	71770.46	58.33417
22.9	71762.61	58.31597
23.2	68970.53	64.63349
23.2	68958.33	64.63493
23.2	68987.85	64.59564
23.2	68966.29	64.75259
23.2	68971.18	64.71535
23.2	68974.03	64.70268
23.2	68971.17	64.72641
23.2	68971.92	64.728
23.2	68908.04	64.87247
23.2	68904.55	64.87391
23.2	68889.28	64.86189
23.2	68866.26	64.85027
22.8	66338.18	69.86868
22.8	66301.06	69.88139
22.8	66269.73	69.87168
22.8	66258.86	69.86871
22.8	66157.19	69.79572
22.8	66164.02	69.79405
22.8	66171.57	69.78311
22.8	66168.39	69.78102
22.8	66161.52	69.78444
22.8	66162.82	69.78257
22.8	66083.7	69.67013
22.8	66095.78	69.65536
22.8	66075.81	69.64879
22.8	66094.89	69.6325
22.8	66111.51	69.62177
22.8	66208.35	69.55168
22.8	66183.44	69.55226
22.8	66175.1	69.55796
22.8	66178.06	69.57029
22.8	66179.96	69.56476
22.8	66221.27	69.55494
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22.8	66192.17	69.56658
22.8	66185.52	69.56223
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22.8	66329.64	69.51143

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0.2	51593.55	21.32377
0.2	51587.48	21.32015
0.2	51593.29	21.31536
0.2	51592.67	21.31522
0.2	51596.35	21.31348
0.2	51597.72	21.31398
0.2	51617	21.31421
0.2	51626.48	21.30043
0.2	51614.47	21.31194
0.2	51615.52	21.31177
0.2	51613.86	21.3211
0.2	51612.04	21.31246
0.2	51604.23	21.31087
0.2	51598.85	21.32326
0.2	51603.72	21.31672
0.2	51607.28	21.29951
0.2	49586.58	24.14066
0.2	49581.69	24.14006
0.2	49573.72	24.13294
0.2	49567.73	24.13162
0.2	49569.84	24.12192
0.2	49554.73	24.13807
0.2	49542.32	24.12761
0.2	49548.23	24.11089
0.2	49561.59	24.08613
0.2	49562.85	24.08664

0.2	49560.25	24.09582
0.2	49545.99	24.10265
0.2	49552.3	24.09421
0.2	49551.84	24.09229
0.2	49558.98	24.09635
0.2	49558.1	24.10092
0.2	49555.88	24.114
0.2	49551.75	24.11792
0.2	49494.84	24.11885
0.2	49502.83	24.11589
0.2	49513.39	24.12256
0.2	49479.72	24.15914
0.2	49467.52	24.18286
0.2	49466.18	24.17442
0.2	49477.89	24.14593
0.2	49479.27	24.15168
0.2	49484.04	24.15685
0.2	49481.91	24.1595
0.2	49510.03	24.15009
0.2	49506.76	24.14738
0.2	49500.78	24.15768
0.2	47870.58	26.65799
0.2	47873.37	26.65204
0.2	47880.99	26.66659
0.2	47892.89	26.65647
0.2	47904.16	26.65666
0.2	47910.6	26.66573
0.2	47910.2	26.6727
0.2	47974.16	26.63401
0.2	47966.89	26.63865
0.2	47963.05	26.63713
0.2	47962.51	26.63669
0.2	47968.28	26.6324
0.2	47975.95	26.65757
0.2	47935.85	26.64183
0.2	47929.97	26.66112
0.2	47928.67	26.63811
0.2	45904.27	30.34152
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0.2	45991.49	30.33323
0.2	46015.79	30.2735
0.2	44102.38	34.59675
0.2	44106.44	34.60457
0.2	44110.98	34.60628
0.2	44116.5	34.6021
0.2	44123.47	34.5923
0.2	44127.21	34.59272
0.2	44134.18	34.58005

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0.2	44296.52	34.48866
0.2	44300.99	34.48162
0.2	44354.03	34.43549
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0.2	44379.32	34.38421
0.2	44378.63	34.4165
0.2	44391.63	34.38276
0.2	44400.67	34.3754
0.2	44397.16	34.3769
0.2	44396.5	34.38204
0.2	44396.65	34.38799
0.2	44401.85	34.39267
0.2	43215.13	37.35114
0.2	43219.91	37.32419
0.2	43223.34	37.32241
0.2	43228.46	37.32672
0.2	43223.06	37.34987
0.2	43226.91	37.34345
0.2	43241.69	37.29119
0.2	43246.28	37.32461
0.2	43302.14	37.26021
0.2	43307.19	37.22968
0.2	43310.91	37.22387
0.2	43312.12	37.24603
0.2	43313.1	37.25205
0.2	43316.48	37.24037
0.2	43323	37.21109
0.2	43387.4	37.13812
0.2	43393.99	37.15321
0.2	43406.48	37.12127
0.2	43403.25	37.14037
0.2	43407.55	37.12443
0.2	43417.41	37.10829
0.2	43441.31	37.09742
0.2	43466.38	36.97859
0.2	43455.73	36.96954
0.2	43451.48	36.98417
0.2	43456.13	36.97828
0.2	43469.68	36.96176
0.2	43475.75	36.96867
0.2	43485.94	36.95021
0.2	43485.48	36.97147
0.2	43490.76	36.95883
0.2	43499.45	36.92958
0.2	43503.71	36.92396
0.2	42356.82	40.17038
0.2	42361.47	40.15714
0.2	42379.45	40.11264
0.2	42381.28	40.11523

0.2	42378.87	40.10674
0.2	42380.89	40.10566
0.2	42386.18	40.10933
0.2	42392.37	40.11511
0.2	42404.2	40.08143
0.2	42491.12	40.00051
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0.2	42500.02	40.0091
0.2	42529.06	39.95227
0.2	42539.56	39.92906
0.2	42552.84	39.88904
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0.2	41261.67	44.3121
0.2	41327.88	44.24748
0.2	41314.94	44.25627
0.2	41321.74	44.1841
0.2	41320.54	44.22604
0.2	41322.2	44.2536
0.2	41336.21	44.24488
0.2	41331.22	44.19751
0.2	39367.21	53.24604
0.2	39373.87	53.21086
0.2	39381.39	53.17546
0.2	39407.26	53.16335
0.2	39415.76	53.15423
0.2	39423.79	53.14825
0.2	39426.87	53.10758
0.2	39443.39	53.02645
0.2	38165.02	62.58233
0.2	38074.7	62.58419
0.2	38014	62.65256
0.2	37972.52	62.67627
0.2	37943.37	62.66215
0.2	37073.76	68.86051
0.2	37022.98	68.8624
0.2	36992.7	68.84847
0.2	37043.9	68.44355
0.2	37029.63	68.4607
0.2	37226.3	67.97416
0.2	37234.56	67.9753
0.2	36005.9	73.52726
0.2	35997.08	73.50313
0.2	36105.68	73.22379
0.2	36118.62	73.18115
0.2	36128.3	73.19153
0.2	36141.17	73.13187
0.2	36167.22	73.08036
0.2	36175.88	73.07635
0.2	36180.53	73.10065
101.7	267194.1	17.95663
101.7	267450.7	17.97437

101.7	267338	17.96541
101.7	267087.1	17.96264
101.7	267624	18.00921
101.7	267376.2	18.01548
101.7	267185.3	18.20529
101.7	267051.6	18.21159
101.7	266916.5	18.21673
101.7	267083.2	18.22369
101.7	267078.2	18.22515
101.7	266741.7	18.22763
101.7	267068.6	18.22117
101.7	267159.3	18.21886
101.7	264539	18.59816
101.7	264634.3	18.61547
101.7	264167.1	18.6966
101.7	263883.3	18.71303
101.7	264093.6	18.71699
101.7	264187.3	18.70185
101.7	259442.8	19.61721
101.7	259475.9	19.63667
101.7	259463.4	19.62964
101.7	259531.3	19.68233
101.7	259413.2	19.68006
101.7	259234.9	19.67042
101.7	259533.1	19.78472
101.7	259484.6	19.80088
101.7	259803.4	19.86148
101.7	259732.4	19.86293
101.7	259702.1	19.86669
101.7	260394.6	19.854
101.7	260333.2	19.85698
101.7	260195.6	19.86441
101.7	260436.4	19.91672
101.7	260485.4	19.96619
101.7	260072.7	19.9762
101.7	260075.7	20.10805
101.7	259979.8	20.11074
101.7	259783.7	20.10667
101.7	260156.9	20.20049
101.7	259959.1	20.21427
101.7	260151.5	20.20612
101.7	259965.3	20.29332
101.7	259953.6	20.29185
101.7	259967.3	20.2954
102.1	256937.5	20.58362
102.1	256810.2	20.58289
102.1	256905.3	20.59137
102.1	257312.9	20.62269
102.1	257206.3	20.61901
102.1	257321.6	20.62061
102.1	256944.2	20.65355

102.1	255503.2	20.92332
102.1	254787.7	21.0255
102.1	254664.2	21.0407
102.5	254265.6	21.16027
102.5	254284.4	21.15612
102.5	254430.9	21.2404
102.5	254156.2	21.26509
102.5	254020.5	21.29293
102.5	254051.8	21.30697
102.5	253837.4	21.324
102.5	253688.8	21.32878
101.7	215140.8	35.56367
101.7	215086.6	35.58872
101.7	215116.7	35.59006
101.7	215189.2	35.59805
101.7	215294	35.65733
101.7	215165.3	35.66162
101.7	214869	35.69966
101.7	214558.9	35.85122
101.7	214634	35.84708
101.7	214710.6	35.82685
102.3	200714.2	46.79472
102.3	200607.8	46.79051
102.3	200523.5	46.80076
103	199299.3	48.14648
103	199314.7	48.17914
103	199218.1	48.18581
103	198794.6	48.72477
103	198427.5	48.90778
103	198270.9	48.95617
103	198366.5	48.9884
103	198429.3	49.1218
103	198484.9	49.13677
103	198629.2	49.14009
103.3	198950.4	49.17823
103.3	198924.8	49.18356
103.3	198854.9	49.16873
103.5	199181.1	49.26744
103.5	199315.4	49.27092
103.5	199081	49.27636
103.5	199000.2	49.30082
103.5	199042	49.31222
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103.5	199162.9	49.29753
103.5	199071.2	49.2947
103.5	198981	49.2918
103.5	199085.1	49.30334
103.5	199162	49.27929
102	267773.6	16.15226
102	268070.6	16.16024
102	268235.6	16.15804

102	268254.8	16.13002
102	268174.2	16.16889
102	267912.3	16.1926
102	268059.3	16.1813
102	267765.1	16.18111
102	267816.7	16.21816
102	267621	16.25414
102	267600.8	16.23606
102	267670.6	16.24655
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102	267712.3	16.23977
102	267478.3	16.26611
102	267677.5	16.27601
102	267346.8	16.28651
102	267431.5	16.31231
102	267228.3	16.33031
102	267245	16.31847
102	267256.9	16.31767
102	267051.9	16.3289
102	267035.8	16.3407
102	267247.1	16.32165
102	267046.7	16.3399
102	266975.2	16.35986
102	267027.3	16.34714
102	266907.2	16.35761
102	266692.6	16.38338
102	266849.7	16.38855
102	266730.4	16.4054
102	266726.3	16.37857
102	266771.4	16.36959
102	266678	16.36435
102	266793.1	16.34849
102	266760.3	16.37943
102	266724.9	16.39369
102	266597.3	16.38135
102	266418.8	16.40535
102	266567.7	16.41324
102	266425.3	16.38595
102	266481.7	16.37965
102	266255	16.41661
102	266341.7	16.43105
102	266270.6	16.44104
102	266458.8	16.40267
102	266491.5	16.41472
102	266283.6	16.43787
102	266371.5	16.44916
102	266255.1	16.45781
102	266480.1	16.43824
102	266375.5	16.42881
102	266467	16.43921
102	266284.9	16.4289

102	266423.4	16.4408
102	266496.2	16.45234
102	266340.9	16.43306
102	266384.1	16.43998
102	266434.6	16.41062
102	266449.9	16.4181
102	266405.1	16.42583
102	266270.5	16.43345
102	266346.2	16.44948
102	266322.7	16.43273
102	266198.1	16.42108
102	266354.8	16.41718
102	266237.2	16.42723
102	266212.8	16.44495
102	266086.3	16.46175
102	266234.1	16.42487
102	266179.8	16.4332
102	266077.1	16.46555
102	265902.8	16.49582
102	265832.3	16.49299
102	265978.4	16.47933
102	265739	16.50558
102	266028	16.47121
102	266262.1	16.44969
102	265926	16.44568
102	266003.5	16.45973
102	265572.9	16.47642
102	265706.3	16.49068
102	265809	16.47709
102	265825.2	16.46939
102	265859.1	16.45433
102	265771.5	16.48039
102	265736.9	16.48562
102	265746.5	16.49415
102	265807.8	16.49005
102	265703.8	16.49189
102	265815.8	16.47896
102	265701.8	16.48286
102	265795.2	16.48113
102	265674.5	16.48719
102	265734.1	16.48932
102	265678	16.48391
102	265529	16.50682
102	265660.5	16.50979
102	265469.8	16.49314
102	265357.2	16.50744
102	265404.9	16.49482
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102	265602.2	16.50011
102	265532.6	16.50619
102	265301.1	16.51921

102	265587.3	16.48485
102	265684.6	16.49532
102	265651.8	16.4665
102	265605.4	16.49599
102	265581.1	16.50193
102	265652.7	16.49556
102	265638.5	16.49424
102	265598	16.50696
102	265618.1	16.52661
102	265532.5	16.53229
102	265365.5	16.51515
102	265348.2	16.51962
102	265486	16.5246
102	265395.1	16.52478
102	265188.6	16.53394
102	265475.1	16.52304
102	265436.4	16.5126
102	265466.7	16.52509
102	265271.3	16.53785
102	265551.8	16.52213
102	265419.6	16.52726
102	265179.7	16.57052
102	265087.7	16.55342
102	265253.4	16.54152
102	265305	16.53204
102	265216.4	16.53081
102	265217.7	16.53735
102	265143	16.56012
102	265095.3	16.57905
102	264981.9	16.594
102	264838.9	16.60923
102	264878	16.60816
102	264569.6	16.6326
102	264654.8	16.62556
102	264803.9	16.64227
102	264846	16.63047
102	264814.9	16.61096
102	264756.4	16.62459
102	264638.8	16.63023
102	264905.2	16.59338
102	265030.7	16.58167
102	264598.6	16.59002
102	264546	16.61902
102	264373	16.6187
102	264663.2	16.60454
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102	264680	16.61667
102	264985	16.62407
102	264756.4	16.60964
102	264674.9	16.6142
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102	264575	16.60954
102	249528.9	19.57876
102	249489.6	19.60038
102	249400.8	19.55687
102	249466.4	19.53767
102	249566.7	19.56907
102	249583.1	19.54397
102	249532.8	19.58129
102	249506	19.57637
102	249405.5	19.58138
102	249315.7	19.59673
102	249363.3	19.60127
102	249195.7	19.61251
102	249632	19.54461
102	249470.8	19.55109
102	249688.9	19.55985
102	249668	19.55182
102	231409.2	23.70547
102	231518.3	23.70712
102	231524.7	23.6718
102	231367.3	23.72954
102	231563	23.63637
102	231425.3	23.6505
102	231485.4	23.69226
102	231473.2	23.67656
102	231437	23.67127
102	231443.8	23.70713
102	231561.6	23.7018
102	231518.8	23.72689
105.6	212624.3	30.53915
105.6	212744.2	30.51731
105.6	212791.8	30.50195
105.6	212870.6	30.49812
105.6	213018.8	30.47436
105.6	212931.7	30.499
105.6	213023.2	30.48598
105.6	213131	30.46199
105.6	213220	30.46072
105.6	213191.9	30.4899
105.6	212831	30.61423
105.6	212807.9	30.62203
105.6	212817.3	30.61136
105.6	212871.4	30.57093
105.6	212938.1	30.53522
105.6	212914.9	30.57368
105.6	212927.7	30.57102

105.6	213051.1	30.55923
105.6	213009.2	30.55524
105.6	213088.3	30.49004
105.6	213104.8	30.50318
105.6	212994	30.53975
105.6	212889.9	30.62439
105.6	212879.8	30.59998
106.8	198012.6	38.65194
106.8	198050.1	38.64839
106.8	198019.3	38.67593
106.8	198160.8	38.621
106.8	198146.9	38.63102
106.8	198279.4	38.62233
106.8	198295.2	38.59911
106.8	198387.6	38.59399
106.8	198402.1	38.61886
106.8	198171.8	38.7674
106.8	198197.5	38.73422
106.8	198181	38.74103
106.8	198219.5	38.72436
106.8	188015.5	47.48118
106.8	188137.9	47.47776
106.8	188094.3	47.45643
106.8	188114.5	47.40102
106.8	188231.9	47.37072
106.8	188270.7	47.44305
106.8	188028	47.68416
106.8	188094.3	47.63738
106.8	188063.3	47.62926
106.8	169418.7	70.85093
106.8	169373.9	70.95664
106.8	169337.1	70.98659
106.8	169337.7	71.08097
106.8	169345.4	71.13516
108.4	169366	71.12434
108.4	169411.2	71.12306
108.4	169458.8	71.15645
108.4	169436	71.25467
108.4	169469.8	71.31325
108.4	169484.7	71.35488
108.4	169544.8	71.31862
108.4	169509.7	71.35429
108.4	169530.9	71.34972
108.4	169499.6	71.41596
108.4	169422.9	71.45606
108.4	169423.9	71.52168
108.4	169404.5	71.56681
108.4	169421.1	71.58223
108.4	169449.7	71.63256
108.4	169531.4	71.64051

Ozone Data (Oz_0RH.xls)

ozone	Rs	1/Poz	1/(1/R2-1/R1)
0	69700	#DIV/0!	#DIV/0!
40.6	169186	243125.3393	-118532
82.25	260969	120010.8058	-95099.3
164.5	370975	60005.40289	-85825.1
246	444245	40125.56413	-82670.6

69700	**Rs**	**time**	**delRs/satdelRs**	
	169308.8	0	0.633164	0.634886
	174023.3	10	0.645166	0.646921
	189832.2	20	0.681063	0.682915
	205744.5	30	0.711624	0.71356
	217579.5	40	0.731456	0.733445
	225552.9	50	0.743643	0.745665
	231036.9	60	0.751537	0.753581
	234912.9	70	0.756894	0.758952
	237868.2	80	0.760861	0.762931
	240157.9	90	0.763868	0.765945
	242128.6	100	0.76641	0.768494
	243784.7	110	0.768515	0.770605
	245176.5	120	0.770261	0.772356
	246383	130	0.77176	0.773858
	247419.8	140	0.773035	0.775138
	248342.3	150	0.774162	0.776267
	248968	160	0.774921	0.777028
	249745.2	170	0.775858	0.777968
	250420	180	0.776668	0.77878
	250981.4	190	0.777338	0.779452
	251460.4	200	0.777907	0.780022
	251884.8	210	0.778409	0.780526
	252383	220	0.778997	0.781116
	252783.2	230	0.779468	0.781588
	253146	240	0.779893	0.782014
	253472.6	250	0.780275	0.782397
	253777.1	260	0.78063	0.782753
	254054.6	270	0.780953	0.783077
	254282	280	0.781217	0.783342
	254562.2	290	0.781542	0.783667
	254837.6	300	0.78186	0.783986
	255042.2	310	0.782096	0.784223
	255226.2	320	0.782308	0.784436
	255437.6	330	0.782552	0.78468
	255595.9	340	0.782733	0.784862
	255781.7	350	0.782947	0.785076
	255994.1	360	0.78319	0.78532
	256126.6	370	0.783342	0.785472
	256253.4	380	0.783487	0.785617

256415.5	390	0.783672	0.785803
256577.5	400	0.783856	0.785988
256681.7	410	0.783975	0.786107
256823.7	420	0.784137	0.786269
256901.8	430	0.784225	0.786358
257037.1	440	0.784379	0.786512
257129.1	450	0.784483	0.786617
257181.4	460	0.784543	0.786676
257257.7	470	0.784629	0.786763
257437.3	480	0.784833	0.786967
257520.5	490	0.784927	0.787061
257664.5	500	0.78509	0.787225
257719.9	510	0.785152	0.787287
257789.4	520	0.785231	0.787366
257843.8	530	0.785292	0.787428
257930.4	540	0.78539	0.787526
257972.6	550	0.785437	0.787573
257999.1	560	0.785467	0.787603
258012.6	570	0.785482	0.787619
258097.2	580	0.785578	0.787714
258152.8	590	0.78564	0.787777
258185.5	600	0.785677	0.787814
258213.9	610	0.785709	0.787846
258269	620	0.785771	0.787908
258292.2	630	0.785797	0.787934
258342	640	0.785853	0.78799
258317.7	650	0.785826	0.787963
258415.1	660	0.785935	0.788073
258476.9	670	0.786005	0.788142
258518	680	0.786051	0.788188
258586.4	690	0.786128	0.788265
258597.4	700	0.78614	0.788278
258681.5	710	0.786234	0.788372
258745.5	720	0.786306	0.788444
258795.4	730	0.786362	0.7885
258782.2	740	0.786347	0.788485
258824.6	750	0.786394	0.788533
258895.6	760	0.786474	0.788613
258848.9	770	0.786422	0.78856
258894.6	780	0.786473	0.788612
258954.2	790	0.78654	0.788679
258954.4	800	0.78654	0.788679
259025.4	810	0.786619	0.788758
259048.9	820	0.786645	0.788785
259122	830	0.786727	0.788867
259109.7	840	0.786713	0.788853
259136.7	850	0.786743	0.788883
259140.2	860	0.786747	0.788887
259116.7	870	0.786721	0.788861
259115.1	880	0.786719	0.788859

259118.7	890	0.786723	0.788863
259118.6	900	0.786723	0.788863
259123.8	910	0.786729	0.788869
259180.6	920	0.786793	0.788932
259141.3	930	0.786749	0.788888
259129.6	940	0.786736	0.788875
259157.9	950	0.786767	0.788907
259161.5	960	0.786771	0.788911
259179.7	970	0.786792	0.788931
259201.9	980	0.786816	0.788956
259245.4	990	0.786865	0.789005
259279.5	1000	0.786903	0.789043
259283.2	1010	0.786907	0.789047
259256.9	1020	0.786878	0.789018

70569.91	0	0.013266	0.013303
76896.91	10	0.100724	0.100998
87338.94	20	0.217351	0.217943
98355.43	30	0.31355	0.314403
108068.4	40	0.382096	0.383135
115942.6	50	0.429237	0.430405
122200.5	60	0.462369	0.463626
127185.9	70	0.48643	0.487753
131211.9	80	0.504526	0.505899
134408.2	90	0.518121	0.51953
137111.5	100	0.529125	0.530564
139393.5	110	0.538081	0.539544
141364.5	120	0.545584	0.547068
143100.7	130	0.552022	0.553523
144665	140	0.55769	0.559207
145997.7	150	0.562423	0.563953
147205.2	160	0.566638	0.568179
148301.6	170	0.570405	0.571956
149295.8	180	0.573774	0.575334
150188.2	190	0.576759	0.578327
150986.9	200	0.579401	0.580977
151726.4	210	0.581822	0.583405
152414.2	220	0.584053	0.585642
153050	230	0.586098	0.587692
153636.4	240	0.587969	0.589568
154196.4	250	0.589742	0.591346
154737.1	260	0.591442	0.59305
155228.1	270	0.592975	0.594588
155692.4	280	0.594416	0.596033
156118.6	290	0.595731	0.597352
156524.3	300	0.596977	0.5986
156921.4	310	0.59819	0.599816
157283.2	320	0.599289	0.600919
157653.7	330	0.60041	0.602043
157995.3	340	0.601439	0.603074

158292.2	350	0.602329	0.603967
158613.7	360	0.60329	0.604931
158915.7	370	0.604188	0.605832
159214.1	380	0.605073	0.606719
159522.9	390	0.605985	0.607633
159783.7	400	0.606753	0.608403
159979.7	410	0.607328	0.60898
160204.6	420	0.607986	0.60964
160456.7	430	0.608722	0.610377
160699.8	440	0.609429	0.611086
160962.2	450	0.61019	0.611849
161176.4	460	0.610809	0.61247
161378.1	470	0.611391	0.613054
161583.2	480	0.611981	0.613645
161793.3	490	0.612584	0.61425
161999.3	500	0.613173	0.614841
162178.7	510	0.613685	0.615354
162334.6	520	0.61413	0.6158
162472.3	530	0.614521	0.616192
162608.2	540	0.614907	0.61658
162744.8	550	0.615294	0.616968
162877.4	560	0.61567	0.617344
163005.3	570	0.616031	0.617706
163157.4	580	0.61646	0.618137
163310	590	0.61689	0.618567
163435.8	600	0.617243	0.618922
163574.7	610	0.617633	0.619313
163695.6	620	0.617971	0.619652
163824.8	630	0.618333	0.620015
163943.7	640	0.618665	0.620347
164061.9	650	0.618995	0.620678
164180.5	660	0.619325	0.621009
164289.2	670	0.619627	0.621312
164408.9	680	0.61996	0.621646
164608.6	690	0.620513	0.622201
164692.1	700	0.620744	0.622432
164691.6	710	0.620743	0.622431
164751.8	720	0.620909	0.622598
164857.8	730	0.621202	0.622891
164970.5	740	0.621513	0.623203
165088	750	0.621836	0.623528
165210.5	760	0.622173	0.623865
165351.6	770	0.622561	0.624254
165459.5	780	0.622857	0.624551
165554.3	790	0.623116	0.624811
165640.4	800	0.623352	0.625047
165743.7	810	0.623634	0.62533
165819.3	820	0.62384	0.625537
165911.2	830	0.624091	0.625788
165992.4	840	0.624312	0.62601

166047.2	850	0.624461	0.626159
166109.9	860	0.624632	0.62633
166180.3	870	0.624823	0.626522
166261.5	880	0.625043	0.626743
166339.5	890	0.625255	0.626955
166411	900	0.625449	0.62715
166453.2	910	0.625563	0.627264
166527.2	920	0.625763	0.627465
166586.3	930	0.625923	0.627625
166661.3	940	0.626126	0.627829
166740.2	950	0.626339	0.628042
166828.9	960	0.626578	0.628282
166917.2	970	0.626816	0.628521
166989	980	0.627009	0.628714
167052.7	990	0.62718	0.628886
167123.5	1000	0.627371	0.629077
167233.2	1010	0.627665	0.629372
167330.1	1020	0.627925	0.629633
167403.2	1030	0.62812	0.629829
167384.8	1040	0.628071	0.629779
167451.4	1050	0.628249	0.629958
167559.6	1060	0.628539	0.630248
167644.6	1070	0.628766	0.630476
167717.8	1080	0.628961	0.630672
167797.7	1090	0.629174	0.630885
167856.1	1100	0.62933	0.631041
167957.4	1110	0.629599	0.631311
168042.1	1120	0.629824	0.631537
168092.1	1130	0.629957	0.63167
168139.6	1140	0.630083	0.631797
168193.2	1150	0.630225	0.631939
168230.7	1160	0.630325	0.632039
168277.3	1170	0.630448	0.632163
168330.2	1180	0.630588	0.632303
168385.5	1190	0.630735	0.63245
168457.5	1200	0.630925	0.632641
168522.6	1210	0.631097	0.632813
168552.2	1220	0.631175	0.632892
168584.9	1230	0.631261	0.632978
168660.1	1240	0.63146	0.633177
168721.6	1250	0.631622	0.63334
168800	1260	0.631828	0.633547
168895.7	1270	0.63208	0.633799
169086.1	1280	0.63258	0.634301

Ozone Data (3RH_6-26.xls)

O3 ppb	Rs	RH	1/R2-1/R1	1/ppb	Poz	1/Poz	1/(1/R2-1/R1)
0	63300	3.8	0	#DIV/0!	0	#DIV/0!	#DIV/0!
0.1	64200	3.8	-2.2E-07	10	1.01641E-08	98385252	-4515400
9.5	87450	3.7	-4.4E-06	0.105263	9.65592E-07	1035634	-229217
26.4	151800	3.7	-9.2E-06	0.037879	2.68333E-06	372671.4	-108576
56.8	253230	3.6	-1.2E-05	0.017606	5.77322E-06	173213.5	-84396.7
85.9	323350	3.6	-1.3E-05	0.011641	8.73098E-06	114534.6	-78708.2
114.6	376000	3.6	-1.3E-05	0.008726	1.16481E-05	85851.01	-76113.8
171.6	455000	3.6	-1.4E-05	0.005828	1.74416E-05	57334.06	-73529.5
229	514900	3.6	-1.4E-05	0.004367	2.32758E-05	42962.99	-72172.7
341	605000	3.6	-1.4E-05	0.002933	3.46597E-05	28851.98	-70696.9

	ave	min	max	std dev
0	63306	63054	63387	68
9.5	87450	87414	87481	22
26.4	151491	151190	151815	172
56.8	253490	252786	255537	782
85.9	323185	322823	323442	158
114.6	376102	375519	376729	287
171.6	454550	453545	455436	563
229	514995	514327	516086	506
341	605130	604555	605569	271

Humidity Data (delRH 0ppb 6-15.xls)

Tc	Power	Rs	Pc	RH	1/delPh	1/(1/R2-1/R1)
25.579	79.999	60750.81	760.69	15.083	365.871	19044600770
25.579	80.000	60679.76	760.69	15.093	327.203	51744227
25.580	80.002	60607.75	760.69	15.104	293.107	25703741
25.586	80.000	57406.08	760.65	16.682	18.130	1042618
25.587	80.000	57383.86	760.65	16.690	18.044	1035339
25.586	80.002	57347.15	760.65	16.702	17.913	1023517
25.585	80.002	57331.68	760.64	16.704	17.893	1018610
25.585	80.001	57311.81	760.65	16.703	17.905	1012375
25.584	80.001	57291.20	760.64	16.703	17.901	1005983
25.583	80.000	57254.49	760.65	16.718	17.748	994783
25.582	80.000	57243.52	760.65	16.717	17.758	991480
25.581	80.000	57207.97	760.65	16.730	17.630	980922
25.580	80.001	57198.17	760.64	16.729	17.638	978049
25.580	80.000	57177.53	760.64	16.721	17.717	972051
25.580	80.000	57162.61	760.64	16.723	17.701	967755
25.579	80.000	57138.13	760.64	16.724	17.695	960787
25.578	80.001	57109.38	760.64	16.732	17.611	952723
25.576	80.000	57097.74	760.64	16.731	17.624	949494
25.477	80.000	54300.87	760.61	18.487	8.800	511436
25.475	79.998	54295.23	760.60	18.491	8.792	510936
25.474	79.997	54295.74	760.60	18.488	8.799	510982
25.473	79.997	54285.63	760.59	18.498	8.776	510088
25.472	79.999	54274.02	760.59	18.500	8.770	509064
25.471	79.998	54252.42	760.59	18.505	8.760	507170
25.469	79.996	54235.94	760.58	18.503	8.764	505734
25.468	79.997	54229.48	760.57	18.496	8.782	505173
25.467	79.998	54215.76	760.56	18.504	8.762	503985
25.466	80.001	54215.52	760.57	18.500	8.772	503964
25.465	80.000	54210.37	760.57	18.506	8.759	503519
25.464	79.999	54197.02	760.57	18.510	8.750	502370
25.463	80.002	54189.42	760.57	18.524	8.716	501718
25.462	79.999	54186.41	760.56	18.524	8.716	501460
25.461	80.000	54190.85	760.56	18.523	8.718	501840
25.460	80.000	54174.25	760.56	18.524	8.716	500421
25.459	80.000	54162.04	760.55	18.526	8.712	499381
25.459	80.000	54149.57	760.56	18.524	8.717	498322
25.458	80.001	54133.02	760.56	18.538	8.683	496924
25.457	80.001	54117.58	760.56	18.546	8.665	495626
25.455	80.000	54098.66	760.56	18.555	8.644	494044
25.454	80.002	54086.26	760.56	18.557	8.640	493012
25.451	80.003	54006.47	760.56	18.638	8.448	486460
25.417	80.001	51590.52	760.50	21.345	4.853	342141
25.416	80.000	51585.06	760.50	21.364	4.839	341901
25.414	79.999	51581.51	760.50	21.356	4.846	341745
25.414	79.997	51586.72	760.50	21.345	4.855	341974
25.413	80.000	51577.96	760.49	21.350	4.851	341589
25.413	79.998	51587.48	760.49	21.335	4.863	342008
25.412	80.003	51587.00	760.49	21.340	4.859	341986

25.410	80.007	51585.72	760.48	21.348	4.854	341930
25.409	80.000	51600.30	760.48	21.332	4.866	342572
25.408	80.000	51604.27	760.48	21.336	4.863	342747
25.408	79.999	51609.45	760.47	21.320	4.875	342975
25.407	80.000	51601.27	760.48	21.329	4.869	342614
25.406	79.998	51593.55	760.47	21.324	4.873	342274
25.406	80.000	51587.48	760.46	21.320	4.876	342007
25.405	80.002	51593.29	760.46	21.315	4.880	342263
25.404	80.001	51592.67	760.46	21.315	4.880	342236
25.403	80.002	51596.35	760.46	21.313	4.882	342397
25.403	80.001	51597.72	760.46	21.314	4.882	342458
25.401	80.001	51617.00	760.46	21.314	4.882	343309
25.401	79.999	51626.48	760.46	21.300	4.893	343729
25.400	79.998	51614.47	760.46	21.312	4.884	343197
25.397	80.000	51615.52	760.45	21.312	4.885	343243
25.398	79.997	51613.86	760.45	21.321	4.878	343170
25.397	79.998	51612.04	760.45	21.312	4.885	343090
25.396	80.000	51604.23	760.44	21.311	4.886	342745
25.395	79.998	51598.85	760.44	21.323	4.877	342508
25.394	80.003	51603.72	760.44	21.317	4.882	342723
25.393	80.003	51607.28	760.44	21.300	4.896	342880
25.392	79.995	51599.99	760.44	21.316	4.884	342558
25.369	80.005	49523.51	760.40	24.136	3.380	267968
25.370	80.003	49522.51	760.40	24.126	3.384	267938
25.368	80.001	49525.48	760.39	24.119	3.387	268025
25.367	80.001	49535.50	760.38	24.105	3.393	268319
25.367	80.000	49549.13	760.39	24.103	3.393	268719
25.367	80.000	49546.56	760.38	24.096	3.396	268644
25.367	80.002	49538.50	760.39	24.100	3.394	268407
25.365	80.000	49533.94	760.39	24.100	3.395	268273
25.365	80.002	49537.95	760.40	24.081	3.402	268391
25.363	80.000	49547.25	760.40	24.080	3.403	268664
25.364	79.998	49559.53	760.40	24.066	3.408	269026
25.364	80.000	49556.09	760.40	24.073	3.405	268924
25.365	80.001	49558.64	760.40	24.082	3.402	268999
25.363	80.000	49564.08	760.39	24.064	3.409	269160
25.363	80.000	49561.59	760.39	24.086	3.400	269086
25.363	80.000	49562.85	760.39	24.087	3.400	269123
25.362	79.998	49560.25	760.39	24.096	3.397	269047
25.361	80.000	49545.99	760.38	24.103	3.395	268627
25.360	79.998	49552.30	760.38	24.094	3.398	268812
25.360	80.000	49551.84	760.37	24.092	3.399	268799
25.359	79.997	49546.42	760.37	24.099	3.396	268640
25.358	80.000	49548.77	760.37	24.088	3.401	268709
25.359	79.998	49553.11	760.36	24.079	3.404	268836
25.358	79.999	49550.51	760.36	24.068	3.408	268760
25.358	80.000	49557.78	760.36	24.070	3.407	268974
25.359	79.999	49558.53	760.36	24.072	3.406	268996
25.357	79.998	49553.16	760.36	24.088	3.401	268838
25.357	79.995	49546.85	760.36	24.091	3.400	268652
25.355	79.995	49555.22	760.36	24.084	3.403	268898
25.356	79.996	49553.18	760.36	24.078	3.405	268838

25.355	80.000	49561.67	760.36	24.069	3.408	269088
25.356	79.998	49566.85	760.35	24.073	3.407	269241
25.355	79.996	49568.26	760.35	24.070	3.408	269283
25.355	80.003	49571.77	760.35	24.066	3.409	269387
25.355	79.996	49569.93	760.34	24.073	3.407	269332
25.355	80.000	49571.82	760.34	24.091	3.400	269388
25.354	79.990	49581.03	760.35	24.080	3.405	269660
25.353	79.998	49577.49	760.35	24.061	3.412	269556
25.353	80.000	49561.94	760.36	24.069	3.409	269097
25.352	79.999	49558.42	760.36	24.076	3.406	268993
25.351	79.997	49560.04	760.35	24.081	3.405	269041
25.351	79.999	49562.21	760.35	24.087	3.403	269105
25.350	79.999	49558.98	760.35	24.096	3.399	269009
25.350	80.002	49558.10	760.35	24.101	3.397	268983
25.350	80.000	49555.88	760.35	24.114	3.393	268918
25.349	80.000	49551.75	760.35	24.118	3.391	268797
25.349	80.003	49544.85	760.35	24.121	3.390	268594
25.348	80.000	49556.80	760.35	24.107	3.396	268945
25.348	80.001	49556.90	760.35	24.103	3.397	268948
25.348	80.000	49547.32	760.35	24.108	3.395	268666
25.348	80.001	49534.22	760.34	24.118	3.391	268281
25.348	80.001	49517.96	760.34	24.136	3.385	267805
25.347	80.000	49514.93	760.34	24.139	3.384	267717
25.347	80.000	49503.95	760.33	24.148	3.381	267396
25.347	79.999	49493.26	760.33	24.161	3.376	267084
25.347	80.000	49496.11	760.33	24.122	3.390	267167
25.346	79.999	49501.84	760.32	24.106	3.396	267334
25.345	79.999	49497.00	760.32	24.130	3.388	267193
25.345	79.997	49493.41	760.32	24.136	3.385	267089
25.344	79.999	49493.63	760.32	24.126	3.389	267095
25.344	79.999	49496.34	760.32	24.124	3.390	267174
25.343	79.999	49493.76	760.32	24.137	3.385	267099
25.344	79.993	49488.65	760.32	24.125	3.390	266950
25.343	79.997	49483.08	760.32	24.139	3.384	266788
25.342	80.000	49476.22	760.31	24.126	3.389	266589
25.342	80.002	49483.65	760.31	24.116	3.393	266805
25.342	80.003	49494.84	760.31	24.119	3.392	267130
25.342	80.001	49502.83	760.31	24.116	3.393	267363
25.341	80.004	49513.39	760.31	24.123	3.391	267672
25.339	80.004	49524.01	760.31	24.138	3.386	267982
25.339	80.001	49524.01	760.31	24.124	3.391	267982
25.339	80.006	49515.02	760.31	24.118	3.393	267719
25.339	80.008	49517.07	760.30	24.123	3.391	267779
25.337	80.001	49502.27	760.31	24.134	3.387	267347
25.337	79.999	49481.57	760.31	24.137	3.387	266744
25.337	80.006	49475.86	760.31	24.156	3.379	266578
25.337	80.004	49463.74	760.32	24.148	3.383	266227
25.336	80.003	49470.97	760.32	24.149	3.382	266436
25.336	80.002	49488.79	760.32	24.139	3.386	266954
25.338	80.001	49495.16	760.32	24.137	3.386	267140
25.338	79.997	49485.41	760.32	24.156	3.379	266856
25.337	79.995	49479.34	760.32	24.165	3.376	266679

25.336	80.000	49479.72	760.31	24.159	3.379	266690
25.336	79.999	49467.52	760.32	24.183	3.370	266336
25.334	80.000	49466.18	760.32	24.174	3.373	266298
25.334	80.000	49477.89	760.32	24.146	3.384	266637
25.333	79.999	49479.27	760.32	24.152	3.382	266677
25.332	80.000	49484.04	760.32	24.157	3.380	266816
25.334	80.000	49481.91	760.32	24.159	3.379	266754
25.332	79.999	49485.20	760.32	24.144	3.385	266850
25.331	80.000	49493.14	760.32	24.134	3.389	267081
25.330	79.998	49498.49	760.32	24.134	3.389	267237
25.330	79.998	49501.55	760.33	24.131	3.390	267326
25.330	79.997	49512.34	760.33	24.125	3.393	267641
25.329	79.996	49510.17	760.33	24.128	3.392	267577
25.328	79.997	49518.91	760.33	24.122	3.394	267833
25.328	80.000	49518.10	760.33	24.131	3.391	267809
25.328	79.993	49517.00	760.33	24.134	3.389	267777
25.327	79.997	49518.18	760.33	24.140	3.387	267812
25.327	79.994	49526.28	760.32	24.128	3.392	268049
25.327	80.003	49528.91	760.32	24.129	3.392	268126
25.327	79.997	49515.42	760.32	24.136	3.389	267731
25.326	80.003	49502.74	760.32	24.150	3.384	267361
25.326	80.004	49495.70	760.32	24.146	3.385	267155
25.325	80.003	49500.22	760.32	24.146	3.386	267287
25.326	80.001	49505.69	760.31	24.157	3.381	267446
25.324	79.999	49504.01	760.31	24.152	3.384	267398
25.323	79.998	49511.03	760.31	24.149	3.385	267603
25.323	80.000	49514.91	760.31	24.150	3.384	267716
25.321	79.997	49506.75	760.31	24.151	3.385	267478
25.321	79.998	49510.03	760.31	24.150	3.385	267573
25.321	80.000	49506.76	760.31	24.147	3.386	267478
25.320	79.999	49500.78	760.31	24.158	3.382	267303
25.320	80.003	49491.89	760.31	24.178	3.375	267044
25.319	80.004	49537.40	760.30	24.158	3.382	268375
25.317	80.001	47905.85	760.25	26.687	2.651	226570
25.317	80.000	47930.84	760.24	26.673	2.654	227130
25.318	80.004	47911.67	760.24	26.685	2.651	226700
25.318	80.002	47903.78	760.24	26.681	2.652	226524
25.318	79.996	47905.22	760.23	26.678	2.653	226556
25.319	80.000	47902.21	760.23	26.677	2.653	226489
25.319	80.002	47923.51	760.22	26.644	2.660	226966
25.321	80.002	47927.20	760.22	26.653	2.658	227048
25.322	79.998	47924.99	760.21	26.674	2.653	226999
25.323	80.003	47931.53	760.21	26.678	2.652	227146
25.324	80.003	47936.45	760.21	26.668	2.654	227256
25.324	80.003	47940.89	760.20	26.653	2.657	227356
25.324	80.004	47944.83	760.21	26.662	2.655	227445
25.325	80.004	47970.54	760.20	26.651	2.658	228025
25.326	80.005	47986.56	760.20	26.633	2.662	228387
25.326	80.004	47974.16	760.19	26.634	2.662	228106
25.326	80.001	47966.89	760.19	26.639	2.660	227942
25.326	80.000	47963.05	760.19	26.637	2.661	227855
25.327	79.999	47962.51	760.19	26.637	2.661	227843

25.327	80.002	47968.28	760.19	26.632	2.662	227974
25.328	80.003	47975.95	760.19	26.658	2.656	228147
25.328	80.001	47986.35	760.20	26.649	2.658	228382
25.328	79.993	47962.06	760.20	26.691	2.648	227833
25.329	80.000	47965.59	760.20	26.666	2.654	227913
25.330	80.004	47972.67	760.20	26.635	2.661	228073
25.330	79.998	47970.19	760.19	26.648	2.658	228017
25.331	79.998	47970.67	760.20	26.640	2.659	228027
25.332	80.004	47970.87	760.20	26.693	2.647	228032
25.333	79.994	47981.46	760.20	26.695	2.647	228271
25.332	79.998	47948.99	760.20	26.677	2.651	227538
25.332	79.998	47934.63	760.19	26.667	2.653	227215
25.332	80.006	47943.77	760.19	26.633	2.661	227421
25.333	80.001	47935.91	760.19	26.646	2.657	227244
25.334	79.999	47935.85	760.20	26.642	2.658	227243
25.334	80.005	47929.97	760.20	26.661	2.654	227111
25.335	80.000	47928.67	760.20	26.638	2.659	227081
25.334	80.002	47928.76	760.19	26.647	2.657	227084
25.335	79.998	47928.81	760.19	26.649	2.657	227085
25.338	80.003	45898.88	760.17	30.362	2.014	187744
25.338	80.002	45906.34	760.18	30.341	2.017	187869
25.339	80.003	45909.67	760.17	30.339	2.017	187925
25.339	80.001	45907.69	760.17	30.336	2.017	187892
25.340	80.001	45896.30	760.17	30.367	2.013	187701
25.340	80.001	45888.45	760.17	30.367	2.013	187570
25.341	80.000	45892.46	760.17	30.367	2.013	187637
25.340	80.002	45891.75	760.17	30.346	2.016	187625
25.339	80.001	45899.80	760.17	30.353	2.015	187760
25.340	80.000	45915.53	760.17	30.317	2.020	188023
25.342	79.997	45921.67	760.16	30.352	2.015	188126
25.342	79.999	45924.44	760.16	30.337	2.017	188173
25.343	80.002	45944.12	760.16	30.310	2.020	188504
25.343	80.001	45955.76	760.16	30.317	2.020	188700
25.341	79.998	45959.19	760.15	30.324	2.019	188758
25.342	80.001	45967.04	760.15	30.315	2.020	188890
25.342	80.006	45976.17	760.15	30.322	2.019	189044
25.343	79.998	45979.61	760.16	30.361	2.014	189103
25.343	80.002	45991.49	760.16	30.333	2.017	189304
25.344	79.999	46015.79	760.15	30.274	2.025	189716
25.343	80.004	46014.31	760.15	30.316	2.020	189691
25.346	79.998	44134.18	760.15	34.580	1.579	161354
25.347	79.999	44142.69	760.15	34.582	1.579	161468
25.347	80.000	44154.29	760.15	34.554	1.581	161623
25.346	80.000	44158.99	760.15	34.564	1.581	161686
25.347	80.000	44169.75	760.15	34.538	1.583	161831
25.347	80.000	44176.98	760.15	34.539	1.583	161928
25.347	80.000	44188.39	760.15	34.542	1.583	162081
25.348	80.000	44195.72	760.15	34.539	1.583	162180
25.347	80.000	44210.34	760.15	34.525	1.584	162377
25.348	80.000	44225.59	760.14	34.521	1.584	162583
25.349	80.000	44230.58	760.15	34.495	1.586	162650
25.349	80.000	44227.84	760.14	34.515	1.585	162613

25.348	80.000	44230.21	760.15	34.522	1.584	162645
25.349	80.001	44234.99	760.14	34.526	1.584	162710
25.349	80.000	44244.26	760.13	34.509	1.585	162835
25.349	80.001	44248.98	760.13	34.529	1.583	162899
25.349	80.001	44260.64	760.13	34.495	1.586	163058
25.349	80.001	44273.70	760.14	34.487	1.587	163235
25.350	80.002	44278.27	760.14	34.498	1.586	163297
25.350	80.000	44289.00	760.14	34.472	1.588	163443
25.349	80.001	44291.22	760.14	34.503	1.585	163473
25.350	79.998	44296.52	760.14	34.489	1.587	163546
25.350	80.002	44300.99	760.14	34.482	1.587	163607
25.351	80.001	44306.99	760.14	34.470	1.588	163688
25.351	80.000	44307.89	760.14	34.454	1.589	163701
25.351	80.002	44304.06	760.14	34.476	1.587	163648
25.351	80.003	44306.71	760.14	34.489	1.586	163685
25.351	80.001	44317.59	760.13	34.464	1.588	163833
25.352	79.999	44320.72	760.14	34.463	1.588	163876
25.352	80.002	44331.76	760.14	34.435	1.591	164027
25.353	79.999	44332.85	760.14	34.442	1.590	164042
25.353	80.003	44340.09	760.14	34.452	1.589	164141
25.353	80.001	44344.57	760.14	34.437	1.590	164202
25.353	79.999	44345.54	760.14	34.456	1.589	164216
25.354	79.999	44346.36	760.14	34.465	1.588	164227
25.353	79.996	44349.36	760.14	34.460	1.589	164268
25.354	79.998	44354.03	760.14	34.435	1.591	164332
25.354	80.001	44369.58	760.14	34.394	1.594	164546
25.355	79.999	44379.32	760.14	34.384	1.595	164680
25.355	79.998	44378.63	760.13	34.417	1.592	164671
25.353	79.997	44391.63	760.13	34.383	1.595	164850
25.354	80.003	44400.67	760.13	34.375	1.595	164974
25.355	80.000	44397.16	760.13	34.377	1.595	164926
25.357	79.999	44396.50	760.13	34.382	1.595	164917
25.357	80.001	44396.65	760.13	34.388	1.594	164919
25.357	80.002	44401.85	760.13	34.393	1.594	164991
25.357	80.003	44410.87	760.13	34.392	1.594	165115
25.357	80.006	44416.03	760.13	34.388	1.594	165187
25.356	79.998	44423.08	760.12	34.371	1.596	165284
25.356	79.999	44434.18	760.13	34.341	1.598	165438
25.357	80.000	44442.72	760.12	34.335	1.598	165556
25.374	79.999	43270.40	760.13	37.282	1.386	150379
25.376	80.001	43279.75	760.13	37.253	1.387	150492
25.377	80.001	43289.03	760.14	37.236	1.388	150604
25.378	80.002	43297.14	760.13	37.238	1.388	150703
25.379	80.002	43297.84	760.14	37.270	1.386	150711
25.381	80.001	43302.83	760.14	37.261	1.386	150772
25.383	80.004	43302.14	760.14	37.260	1.386	150763
25.384	80.004	43307.19	760.13	37.230	1.388	150825
25.386	80.001	43310.91	760.14	37.224	1.388	150870
25.388	80.002	43312.12	760.13	37.246	1.387	150884
25.388	80.000	43313.10	760.13	37.252	1.386	150896
25.389	79.999	43316.48	760.13	37.240	1.387	150937
25.390	80.000	43323.00	760.14	37.211	1.389	151017

25.393	80.000	43333.32	760.14	37.192	1.390	151142
25.394	80.003	43332.42	760.13	37.218	1.388	151131
25.396	80.001	43338.21	760.13	37.215	1.388	151201
25.397	80.000	43343.53	760.13	37.216	1.388	151266
25.400	80.000	43351.62	760.13	37.198	1.389	151365
25.401	79.999	43353.63	760.13	37.184	1.390	151389
25.403	80.002	43369.04	760.13	37.181	1.390	151577
25.404	80.000	43372.03	760.13	37.178	1.390	151614
25.405	80.001	43369.22	760.13	37.164	1.391	151580
25.406	80.001	43366.69	760.13	37.184	1.389	151549
25.407	80.000	43376.07	760.13	37.169	1.390	151663
25.409	80.002	43377.20	760.13	37.173	1.390	151677
25.409	79.998	43376.81	760.13	37.147	1.391	151672
25.411	80.001	43382.03	760.14	37.138	1.392	151736
25.413	80.001	43383.71	760.13	37.156	1.390	151757
25.414	80.001	43384.07	760.14	37.141	1.391	151761
25.415	80.002	43387.40	760.14	37.138	1.391	151802
25.416	80.002	43393.99	760.14	37.153	1.390	151883
25.417	80.002	43406.48	760.14	37.121	1.392	152036
25.419	80.001	43403.25	760.14	37.140	1.391	151996
25.421	80.000	43407.55	760.14	37.124	1.392	152049
25.423	80.004	43417.41	760.15	37.108	1.393	152170
25.425	79.997	43441.31	760.15	37.097	1.393	152464
25.427	79.998	43446.02	760.14	37.123	1.391	152522
25.428	80.003	43434.08	760.14	37.077	1.394	152375
25.428	80.005	43427.59	760.14	37.072	1.394	152295
25.429	80.002	43421.69	760.14	37.080	1.394	152223
25.429	80.002	43405.57	760.14	37.098	1.393	152025
25.431	80.000	43427.06	760.13	37.073	1.394	152289
25.432	80.001	43420.56	760.14	37.102	1.392	152209
25.432	80.002	43424.63	760.13	37.087	1.393	152259
25.432	80.000	43432.49	760.14	37.083	1.393	152355
25.434	80.000	43439.72	760.14	37.041	1.396	152444
25.435	80.001	43431.85	760.13	37.033	1.396	152347
25.435	80.000	43416.17	760.13	37.051	1.395	152155
25.437	80.001	43417.17	760.14	37.027	1.397	152167
25.437	80.001	43415.86	760.14	37.035	1.396	152151
25.437	80.002	43420.30	760.13	37.042	1.395	152205
25.437	80.002	43436.07	760.13	37.010	1.398	152399
25.438	79.999	43455.89	760.13	37.013	1.397	152644
25.439	80.001	43481.79	760.13	36.978	1.399	152964
25.439	79.999	43482.27	760.13	36.990	1.399	152970
25.440	79.999	43481.76	760.14	37.004	1.398	152963
25.440	80.001	43466.38	760.14	36.979	1.399	152773
25.441	80.000	43455.73	760.14	36.970	1.400	152642
25.443	80.002	43451.48	760.14	36.984	1.399	152589
25.444	80.003	43456.13	760.14	36.978	1.399	152647
25.444	80.000	43469.68	760.14	36.962	1.400	152814
25.445	80.001	43475.75	760.14	36.969	1.400	152889
25.445	79.999	43485.94	760.14	36.950	1.401	153015
25.447	80.003	43485.48	760.14	36.971	1.399	153009
25.448	79.999	43490.76	760.14	36.959	1.400	153075

25.449	79.999	43499.45	760.14	36.930	1.402	153182
25.449	79.998	43503.71	760.14	36.924	1.402	153235
25.450	80.000	43502.48	760.14	36.933	1.401	153220
25.451	80.000	43502.93	760.14	36.915	1.402	153226
25.452	80.000	43500.21	760.14	36.910	1.403	153192
25.463	80.001	42464.76	760.15	40.016	1.228	141078
25.463	79.999	42468.46	760.15	40.025	1.227	141118
25.463	80.001	42473.79	760.15	40.001	1.228	141177
25.465	79.996	42478.24	760.15	39.987	1.229	141226
25.465	79.999	42476.80	760.15	40.024	1.227	141211
25.465	79.997	42481.54	760.15	40.027	1.227	141263
25.464	79.999	42484.45	760.14	40.014	1.228	141295
25.463	79.999	42491.12	760.14	40.001	1.228	141369
25.464	79.999	42500.91	760.14	39.970	1.230	141477
25.465	80.003	42500.02	760.15	40.009	1.228	141467
25.466	80.000	42507.18	760.15	39.988	1.229	141547
25.466	79.999	42496.82	760.14	40.010	1.228	141432
25.467	80.001	42498.13	760.14	39.969	1.230	141446
25.468	79.997	42502.88	760.14	39.941	1.231	141499
25.468	79.998	42504.83	760.14	39.972	1.230	141521
25.468	80.000	42505.76	760.15	39.989	1.229	141531
25.469	80.001	42509.54	760.15	39.935	1.231	141573
25.469	80.001	42522.09	760.15	39.935	1.231	141712
25.470	79.999	42528.34	760.15	39.970	1.229	141782
25.471	80.000	42515.55	760.15	39.975	1.229	141640
25.471	80.000	42509.33	760.15	39.975	1.229	141571
25.472	79.998	42506.07	760.15	39.992	1.228	141534
25.473	79.999	42511.34	760.14	39.955	1.230	141593
25.473	79.998	42519.36	760.14	39.928	1.231	141682
25.475	80.000	42522.73	760.14	39.917	1.232	141719
25.476	80.000	42526.68	760.14	39.939	1.231	141763
25.478	79.998	42533.88	760.15	39.919	1.231	141843
25.478	79.997	42529.06	760.15	39.952	1.230	141790
25.479	80.003	42539.56	760.15	39.929	1.231	141906
25.478	80.002	42552.84	760.15	39.889	1.233	142054
25.479	80.000	42558.40	760.15	39.896	1.232	142116
25.480	80.000	42564.39	760.15	39.895	1.232	142183
25.481	80.002	42572.09	760.15	39.883	1.233	142269
25.483	80.002	42577.95	760.16	39.885	1.233	142335
25.484	80.002	42587.88	760.16	39.857	1.234	142446
25.485	79.999	42593.96	760.17	39.856	1.234	142514
25.484	80.000	42596.75	760.17	39.856	1.234	142545
25.486	80.001	42609.28	760.17	39.810	1.236	142685
25.509	79.999	41289.58	760.16	44.267	1.047	128890
25.510	79.998	41303.46	760.15	44.228	1.048	129025
25.512	79.999	41315.12	760.15	44.231	1.048	129139
25.513	80.001	41322.18	760.16	44.237	1.047	129208
25.513	80.005	41327.88	760.16	44.247	1.047	129264
25.513	80.008	41314.94	760.16	44.256	1.047	129138
25.513	80.010	41321.74	760.16	44.184	1.049	129204
25.513	80.011	41320.54	760.16	44.226	1.048	129192
25.513	80.011	41322.20	760.16	44.254	1.047	129208

25.512	80.006	41336.21	760.15	44.245	1.047	129346
25.513	80.001	41331.22	760.15	44.198	1.049	129297
25.533	80.001	39491.45	760.11	53.030	0.804	112850
25.533	80.001	39507.06	760.10	52.977	0.805	112978
25.533	79.999	39511.19	760.10	52.986	0.805	113012
25.535	79.997	39505.32	760.09	52.989	0.805	112964
25.535	80.001	39509.15	760.09	52.973	0.805	112995
25.535	80.000	39517.35	760.09	52.956	0.806	113062
25.536	80.003	39528.24	760.09	52.936	0.806	113151
25.536	80.001	39545.16	760.09	52.929	0.806	113290
25.536	80.001	39554.65	760.08	52.886	0.807	113368
25.536	80.001	39572.96	760.08	52.892	0.807	113518
25.549	80.002	37946.83	760.07	62.447	0.644	101091
25.550	80.004	37953.86	760.07	62.436	0.644	101141
25.550	80.004	37956.73	760.07	62.467	0.644	101162
25.549	80.000	37960.21	760.07	62.445	0.644	101187
25.551	80.003	37967.32	760.07	62.418	0.644	101237
25.551	80.003	37981.47	760.07	62.386	0.645	101338
25.574	80.000	37088.27	760.08	68.304	0.572	95219
25.574	80.000	37093.67	760.08	68.324	0.572	95255
25.574	80.001	37108.29	760.08	68.265	0.573	95351
25.574	80.000	37119.70	760.08	68.246	0.573	95427
25.575	79.999	37126.95	760.08	68.242	0.573	95475
25.577	80.001	37131.52	760.08	68.261	0.573	95505
25.577	80.000	37144.18	760.08	68.240	0.573	95589
25.577	80.000	37175.54	760.08	68.151	0.574	95797
25.577	80.001	37176.05	760.07	68.172	0.574	95800
25.579	80.000	37181.12	760.08	68.108	0.574	95834
25.579	80.000	37191.06	760.08	68.057	0.575	95900
25.580	80.001	37195.67	760.07	68.012	0.575	95930
25.580	80.001	37203.91	760.07	68.007	0.575	95985
25.581	79.998	37215.89	760.08	67.996	0.575	96065
25.580	80.001	37226.30	760.07	67.974	0.576	96134
25.581	80.001	37234.56	760.07	67.975	0.576	96190
25.581	80.001	37245.19	760.07	67.936	0.576	96261
25.582	80.001	37261.81	760.07	67.856	0.577	96372
25.584	80.000	37284.15	760.07	67.877	0.577	96521
25.585	80.000	37295.99	760.07	67.887	0.577	96601
25.586	79.999	37296.77	760.07	67.802	0.577	96606
25.587	79.999	37300.85	760.07	67.783	0.578	96633
25.616	80.000	36105.68	760.06	73.224	0.523	89001
25.615	80.000	36118.62	760.06	73.181	0.523	89080
25.616	79.999	36128.30	760.05	73.192	0.523	89138
25.617	79.999	36141.17	760.05	73.132	0.524	89217
25.619	79.999	36151.53	760.05	73.129	0.523	89280
25.619	80.000	36158.73	760.05	73.131	0.523	89324
25.620	80.000	36167.22	760.06	73.080	0.524	89376
25.620	79.998	36175.88	760.06	73.076	0.524	89429
25.621	80.000	36180.53	760.05	73.101	0.524	89457

APPENDIX G: Test Equipment and Procedure

Test Equipment:

MiCS 2610 ozone sensor	humidity sensor	bubbler (humidifier)
Teflon tubing – 3 meters	temperature sensor	mass flow meter
Swagelok stainless steel fittings	pressure sensor	ozone generator
2 needle valves	0.5 L/min pump	ozone analyzer
4 T-connectors	glass tube	isolation box
DAC	Heat tape	PC

Test Setup:

Put MiCS ozone sensor, humidity sensor, and temperature sensor inside glass tube and connect to circuit board. Seal glass tube with Teflon tape to make airtight. Attach pump and pressure sensor to back end of glass tube using Teflon tubing. (The pump is placed on the back end and pulls air through so as not to change the ozone concentration. Also note that the humidity and temperature sensors are placed after the ozone sensor in the glass tube for the same reason.) Place inside isolated box, with Teflon tubing exiting through a hole in the box. The isolation box is used to ensure that the sensor is not jarred or otherwise disturbed during testing. Attach circuit board to DAC, which should be connected to a computer running the labVIEW program. Connect mass flow meter in line just in front of the box. Put an ambient dump just before the mass flow meter using one of the T-connectors. If pump is disconnected, put a needle valve on the ambient dump to control the flow rate. Attach T-connector to have a Teflon tubing branch lead to ozone analyzer and one leading to ozone generator. Create humidity loop between ozone generator and ozone analyzer (with bubbler wrapped in heat tape) using two t-connectors and a needle valve (needle valve is placed on the direct line between generator and sensor). Attach zero-air source to generator.

Test Procedure:
Fill bubbler halfway. Open needle valve all the way to create lowest humidity value (approximately 18% RH). If lower humidities are desired, disconnect humidity loop – zero air should have only about 3%RH by itself. Set flow rate on ozone generator to 5000 sccm. To test using constant flow rate, disconnect pump and adjust the flow using the needle valve on the ambient dump. Turn on labVIEW program, which will then begin recording data.

Set ozone generator to produce desired ozone concentration. Allow 15 minutes between changes to ensure steady state. Once system reaches steady state, wait two minutes to obtain steady state data, recording ozone concentration from ozone analyzer each minute. Change relative humidity (by adjusting needle valve) in approximately 5%RH increments from the lowest humidity value to the highest that the system will produce (turning on heat tape will allow higher RH values), waiting 15 minutes between each step. Repeat for different ozone concentrations.

Test Procedure for Calibration

1. Set up system – Set up power to 82.5 mW using labVIEW program, turn on pump, put water in bubbler, turn on ozone generator (set flow rate to 5000sccm)
2. Set ozone generator to 0ppb
    a. Run humidity tests. Start at 20%RH and go to 85%RH in 15%. Wait 20 minutes at each step to ensure steady-state. At higher humidities, use heat tape that is wrapped around a bubbler to increase the %RH. (Generally, I turn it on at 35% with dial set to ~10, then ~20 at 50%, ~30 at 65%, ~40 at 85%. Note that if you set the heat too high, water will condense in the pipes and eventually form drops that travel to the sensor and temporarily screw it up)
    b. Record ozone concentration off of ozone analyzer at steady state (could probably hook analyzer to computer and modify program to record this also). Note that readings are inaccurate for the first two minutes after humidity change.
3. Run step 2 with ozone generator set to 50ppb. Make sure sensor reaches steady state before changing it again.
4. Run step 2 with 100ppb ozone.
5. Run step 2 with 150ppb ozone.

Calibration Procedure

1. Compile data
    a. Look at last 2-5 minutes of data at each calibration point. Filter out any large power fluctuations (should be within $\pm$ .02 of 82.5mW)
    b. Input ozone readings into another column
    c. Put good data in separate file – one long column of data works best if you're using MATLAB.
2. Convert %RH and ozone ppb into pressure (kPa) – this requires the temperature and chamber pressure. (If you're using my lang.m MATLAB program, this is done for you)
3. Fit the data to the following formula to obtain calibration constants:

    resistance = 1/(A*(Ph/bH+C*Poz/Boz)/(1+Ph/bH+Poz/Boz)+1/Ro)

    The program I wrote (lang.m) will find the constants for you.

4. The formula from the previous step has been rearranged and put into the LabVIEW program: the constants just need to be updated for each sensor, which you've obtained from the previous step.

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