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Indices of Water Content in Gaseous Systems, Their Measurement, and Relationship to Each Other^{1,2}

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ABSTRACT

The quantity of water available to microorganisms influences both their growth and death kinetics. The water in microbial cells cannot be measured directly. The equilibrium atmospheric water conditions are usually measured and reported. The attributes of the several measuring units that can be used to indicate the water content in a gaseous atmosphere are described. Equations for their calculation and for converting from the several measurement units described to relative humidity at test temperature are presented.

The survival and growth of microorganisms requires adequate availability of water. Preservation of biological material by drying is accomplished because microorganisms cannot grow and reproduce under conditions of low water availability. Mossel and Ingram (6) in discussing the physiology of microbial spoilage of food list the "physical state of the food" as one of the important intrinsic factors affecting spoilage. They discuss the role of water vapor pressure and then specify the lowest equilibrium relative humidity (ERH) permitting development of spoilage organisms and list the data sources. Minimum ERH values are:

Organism	Minimum ERH
normal bacterial	0.91
normal yeasts	0.88
normal molds	0.80
halophilic bacteria	0.75
xerophilic fungi	0.65
osmophilic yeasts	0.60

They also list "alarm water contents" of some common "dry" foods assuming an RH of 0.70 and temperature of 20 C. For most dry food products the "alarm water content" is 13 to 15%.

Water availability is of critical importance in the preservation of "intermediate moisture foods" (IMF). These are foods that have a water content usually greater than 18% and an ERH of 0.60 to 0.85. The percent water is above the "alarm water content;" therefore, IMF's are

not preserved by the reduced ERH alone. Intermediate moisture foods are preserved by a combination of the bacteriostatic effects of reduced ERH, reduced pH, and the addition of chemical preservatives.

The critical importance of water on microbial growth is universally recognized; however, there is little agreement regarding how to measure and report water conditions.

Bone (3) states that "Water activity is the basic control factor in preservation of dry foods, salted foods, mincemeats, jellies, etc." At a later point in his paper Bone states that "Water activity of a food can be defined as the ratio of the water vapor pressure above the food to the vapor pressure of pure water. A thermodynamic principle says that at equilibrium, the activity of a vapor equals the activity of the corresponding liquid phase. Thus, the measurement of the equilibrium relative humidity of a food is also a measure of its water activity."

The dry heat destruction of bacterial spores is a function of the water content of the spore during heating. Therefore, the D-value or other microbial destruction rate parameter must be related to a water content parameter—either water content itself or the relative vapor pressure of the water in the spore. For a specific spore prepared in a specific manner it is assumed that there is a fixed relationship between spore water content and the relative humidity or other measure of the water in the atmosphere adjacent to the spore. Some systems are not. (Conservative in the context we are using it means that the physical conditions of temperature and pressure of the system can change but the measure will not change.)

Several different indices—relative humidity at test temperature, relative humidity at ambient temperature, parts per million water, absolute humidity and dew point—are being used in the NASA Planetary Quarantine program to quantify water vapor in dry heat microbial destruction systems. To compare dry heat destruction data, where the concentration of water in the atmosphere surrounding the spores has an effect on the spore destruction rate, it is necessary that all the data be in terms of a single water measurement index. Since humidity is a material variable, it cannot be linearly

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converted from one form to another as can physical variables such as temperature and pressure. Furthermore, it is sometimes necessary to specify additional physical parameters before such a conversion can be made.

In this report we will discuss measures of atmospheric water which can be used to describe the equilibrium conditions and indirectly the water availability when microorganisms are dry, are in the intermediate moisture range, or are in the growth range. We will try to relate the different measure of atmospheric water and show how our formula and tables can be used.

DESCRIPTION OF WATER IN BACTERIAL SPORES

Scott (8) used the term "water activity" to describe the status of the water in a solution or substrate. He defined water activity (a_w) as the ratio of the apparent vapor pressure of the solution or substrate to the saturated vapor pressure at a given temperature. In his examples, the a_w of solutes in aqueous solutions are calculated using the osmotic coefficient, the molal concentration of the solute and the number of ions generated by each molecule of solute. Theoretically, under equilibrium conditions the water vapor pressure in the gas above and in the solution will be equal.

The water vapor pressure in the gas can be measured by psychrometric methods. The ratio of the water vapor pressure in the headspace to the saturated water vapor pressure is the relative humidity and, at equilibrium, is theoretically equal to the water activity of the solution.

The concept of water activity of solutions adopted by Scott (8) has been extended to microbial cells and spores. Since it is possible to confuse terms, the following are suggested to clarify the description of water in microbial systems: Relative humidity is a real, engineering unit that is the ratio of two measured quantities-the actual water vapor pressure in a system and the saturated water vapor pressure at the same temperature. It is used with gaseous systems, for example, to describe the water condition in the atmosphere surrounding bacterial cells or spores. Water activity, aw, is a term used to describe the relative water availability inside a microbial cell or spore. Water activity is a theoretical term that cannot be directly measured. If the cell or spore is in equilibrium with the surrounding atmosphere, theoretically the water activity of the spore is equal to the external relative humidity.

When dry microbial cells are heated, the rate of destruction of the cells is influenced by the test system. An "open system" is open to the atmosphere; the ambient atmospheric water vapor pressure conditions determine cell-water content. In a "closed system" the microorganisms are in a fixed volume hermetic container where the total water present initially will be present throughout the test.

In reporting dry heat research data, if in a microbial destruction rate test the relative humidity is measured and controlled, then the results should be reported as a function of relative humidity, not water activity. Whenever a relative humidity value is reported, the temperature at which the relative humidity was measured should be included; for example, 0.2% RH (110 C).

PROPERTIES OF GAS MIXTURES

Before outlining the terms used to describe water content, the physical laws that describe the behavior of gas mixtures will be reviewed.

The physical state of a gas can many times be described by the Ideal Gas Law. The equation of the state of an ideal gas is expressed by the following relationship:

64.1

60.1

P = pressure exerted by the gas
 T = absolute temperature
 R = ideal gas proportionality constant

 $PV \neq nRT$

n = number of moles of gas

V = volume occupied by the n moles

In a mixture of gases, the molecules of each component gas are distributed throughout the containing vessel. The total pressure is the sum of the pressures exerted by the molecules of the component gases. By definition, the *partial pressure* of any one component gas is the pressure that the component gas will exert when it is alone in the same volume at the same temperature. Also by definition, the *pure-component volume* of a constituent gas is the volume that the component gas alone will occupy at the system pressure and temperature.

For an ideal gas, many properties are additive. The total pressure (P) is equal to the sum of the partial pressures.

$$P = p_a + p_b + p_c + \dots$$

$$p_a, p_b, p_c, \text{ etc. are the partial pressures of component gases a, b, c, etc.}$$

This is a statement of Dalton's Law. In the same fashion, the total volume is equal to the sum of the pure-component volumes.

$$V = V_a + V_b + V_c + \dots$$

$$V_a, V_b, V_c, \text{ etc. are pure-component volumes}$$
of gases a, b, c, etc.
[3]

Equation 3 is many times referred to as the Law of AMAGAT or Leduc's Law.

An understanding of these relationships is required to understand the interrelationships between different methods of measuring water in an air atmosphere. For analytical purposes, the mixture of water and air is usually represented as a bi-component mixture of dry air and water vapor where the dry gas is a complex mixture of gases including nitrogen, oxygen, carbon dioxide, and traces of the other gases found in the atmosphere. The molecular weight of the dry gas is taken to be 28.967.

While dry air exhibits the characteristics of an ideal gas, water vapor deviates somewhat from the Ideal Gas Law. However, this deviation is second order in effect and can be neglected.

DESCRIPTION OF WATER IN GAS MIXTURES (Humidity)⁴

There are many distinct humidity forms in use today. We shall describe eight forms that are used to describe the water vapor condition in dry heat or food storage atmospheres. All forms can be divided into two groups:

- I. Expressions based on water substance
 - A. Vapor density
 - B. Relative humidity
 - C. Humidity to gas ratios
 - 1. Specific humidity (mixing ratio)
 - 2. Mole fraction
 - 3. Parts per million
- II. Phenomenological forms
 - A. Vapor pressure
 - B. Dew point
 - C. Wet and dry bulb psychrometry

Vapor density

The gravimetric determination of water vapor in a gas yields data in the form "weight of water per volume of gas" at the temperature of the experiment.

Vapor density is useful because rates of evaporation, diffusion, and reaction are related to differences in concentration of the constituents of a system; also because it directly states the quantity of water vapor by weight in a given volume, rather than as a ratio involving other data.

The numerical value of the vapor density for a particular sample varies with both temperature and pressure in open systems such as we ordinarily encounter in most humidity problems. Therefore, it is sometimes cumbersome to handle in computations.

In a closed system the vapor density method is a conservative form maintaining the same value with changes in temperature and pressure. In the dry heat studies carried out at the FDA-Cincinnati Laboratories, humidity data has been reported using this form (4).

Relative humidity

The relative humidity form is widely used by the comfort air-conditioning industry and by persons involved in the storage and processing of materials. It is unique among the humidity reporting forms because the moisture content of many materials is a direct function of relative humidity; therefore, changes in water content are directly proportional to changes in RH.

Relatve humidity is defined in ASHRAE (2) as the ratio of the mole fraction of water vapor present in the air

to the mole fraction of water vapor present in saturated air at the same temperature and barometric pressure. It approximately equals the ratio of the partial pressure or density of the water vapor in the air to the saturation pressure or density, respectively, of water vapor at the same temperature.

The relative humidity form is not conservative with respect to either temperature or pressure in closed or open systems. Since the relative humidity may be defined as the ratio of the moisture content of a space to the saturation moisture content, it is affected by temperature (the reference saturation moisture content is temperature dependent). Changes in the pressure of a given sample affect the relative humidity. When there is a change in temperature and/or pressure, the corresponding change in relative humidity to the defining equation, (b) adjustment of the individual terms, and (c) recombination to determine the new relative humidity.

Humidity to gas ratios

Three methods of expressing humidity as a ratio will be considered in this discussion:

Specific humidity =	Weight of water vapor				
	Weight of dry gas				
Mole fraction =	Moles water vapor				
Mole fraction -	Total moles wet gas				
X	Volume of water vapor				
Parts per million =	Million (10 ⁶) volumes of wet gas				

Specific humidity, humidity ratio (2), humidity (9), mixing ratio, and absolute humidity (7) are all terms used to describe the ratio of the weight of water vapor to a unit weight of coexisting dry gas. The dry basis ratio is used principally because it is a completely conservative form and lends itself readily to computations where the dry gas fraction does not change.

Mole fraction is the moles of water vapor to the total moles of wet gas. It is an equally conservative form.

Parts per million (ppm) is the ratio of the volume of the water vapor to a million volumetric parts of total gas. Using the Ideal Gas Law, the ppm of water can be related to the partial pressure of water vapor (p_w) in the following manner:

$$ppm = \frac{10^6 p_w}{P}$$
 [4]

 $p_w = partial pressure of water vapor P = total pressure$

Since it is based on pure-component volume, it also represents moles of water per million total moles of gas and, therefore, a million-fold measure of the mole fraction of water. This measure is also independent of temperature and total pressure barring any phase change. Although this measure is usually employed to

⁴In preparing this section of the report, "Humidity Reporting Forms and Humidity Sensors" by Elias J. Amdur (1) has been used extensively.

indicate the water content of relatively dry gases, it can be used at all water levels up to saturation.

Vapor pressure

The vapor pressure of water is an absolute humidity form and the rate of evaporation, diffusion, and reaction are proportional to vapor pressure differences as well as to vapor density. The vapor pressure is, in fact, the preferred form for most purposes, largely because it is more conservative. A numerical value for the vapor pressure varies with total pressure in open systems but is not affected by temperature. In closed systems, the value of the vapor pressure depends on both total pressure and temperature.

Dew point

The water vapor pressure in a system consisting of a vapor space and a plane water surface in isothermal equilibrium is known as the "saturated" vapor pressure of water at that temperature. The saturation vapor pressure of water as a function of temperature is very accurately known. Because of this relationship, it is theoretically possible to determine the vapor pressure in a space by determining the temperature of a plane water surface which is in equilibrium with it.

The dew point determination, however conducted, is an experiment designed to approximate the saturation temperature of the vapor in a space. In the classical procedure one cools a surface until dew forms on it and determines the temperatures at which the dew just forms and just evaporates from the surface. The mean of these two temperatures is regarded as the dew point.

In spite of theoretical difficulties which prevent attainment of high precision, the dew point measurement yields a useful approximation of the saturation temperature and, therefore, the dew point form is a useful alternative method of expressing the absolute humidity. It is also very useful in itself because it indicates the temperature at which moisture will condense on surfaces or clouds will form in a space. This is a uniquely useful property of this reporting form.

Since the classical dew point determination involves bringing a surface to a "dew point" temperature, this type of measurement is independent of the original temperature of the gas being investigated. Since compression or expansion of a gas mixture affects the water vapor pressure, the dew point is also affected by total pressure changes. The dew point and vapor pressure are therefore equally conservative.

Wet and dry bulb psychrometry

The wet and dry bulb psychrometric instrument in the form of the sling psychrometer is probably the most widely used humidity measuring instrument. It is ordinarily considered to be a relative humidity instrument, but examination of the simplified basic equation for this instrument indicates that it yields absolute humidity:

$$(\mathbf{e}_{w} - \mathbf{e}_{a}) = \mathbf{C}(\mathsf{T}_{a} - \mathsf{T}_{w})$$
[5]

 $e_w =$ the vapor pressure of the water on the wick $e_a =$ the unknown ambient vapor pressure $T_a =$ the dry bulb temperature $T_w =$ the wet bulb temperature

The two temperatures, T_a and T_w , are read from the instrument. The value of e_w can be determined from a vapor pressure table knowing T_w . Thus, e_a is the only unknown variable. The percent relative humidity may be obtained by dividing e_a by e_w and multiplying by 100. In practice, tables, charts, or accepted empirical equations of RH vs. the wet and dry bulb are generally used.

The constant C is the ratio of the sensible heat transfer coefficient in the particular bulb design used to the vapor transfer coefficient converted to its equivalent value in terms of heat of evaporation. Since this ratio is not a constant unless the air velocity exceeds 900 ft/sec, a motorized blower drawing the air sample over the bulbs yields more satisfactory data than are obtained with a sling instrument.

It should also be noted that C is proportional to the total pressure as found from both theoretical and experimental considerations. Thus for measurements made at pressures significantly different than one atmosphere, normal psychrometric charts should not be used. Special nomographic charts for the appropriate pressure should be consulted (9).

Wet and dry bulb psychrometry is primarily used as a method of measurement rather than as a reporting form for water vapor in air. Since this method is a measure of the vapor pressure of water, it retains the conservative qualities of vapor pressure measurements.

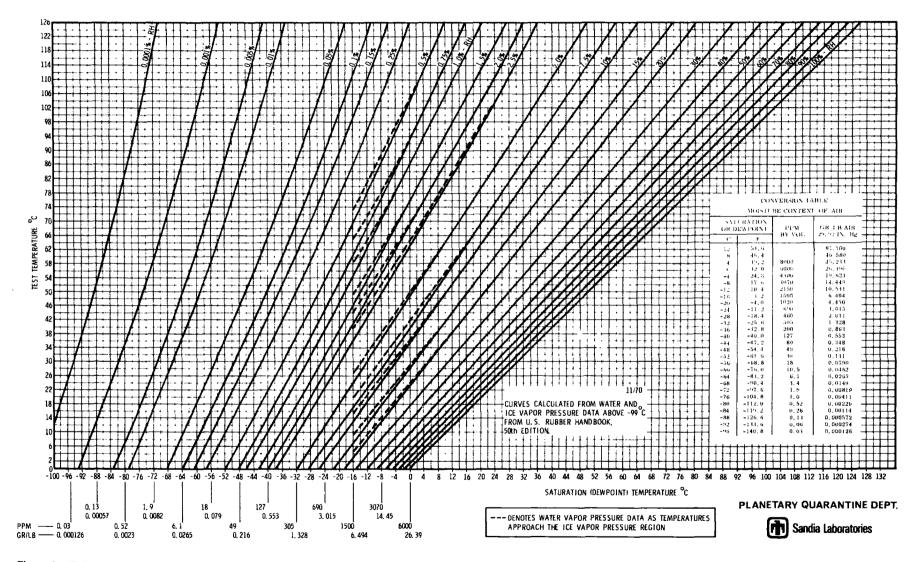
WATER MEASUREMENT CONVERSION TO RELATIVE HUMIDITY

Relative humidity is the form of choice for reporting the water vapor characteristics of the gaseous environments surrounding microorganisms during dry heat testing or food product storage, since it is equal to the water activity when the cell and surrounding atmosphere are in equilibrium. During non-equilibrium periods, relative humidity again appears to be the proper choice from a diffusional transport viewpoint. Thus, it is desirable to have equations at hand which relate other water measurements to relative humidity.

Our general plan is to discuss water measurement conversion in the same order that we discussed the measurement forms. However, since vapor pressure is basic to other calculations we shall discuss it first, followed by a short discussion of the effect of temperature on relative humidity, before proceeding to the other forms.

Vapor pressure to relative humidity

The ratio of the partial pressure of the water vapor to the saturated water vapor pressure at the same temperature is the *relative humidity* of the gas. Relative humidity is thus expressed as



[6]

$$\mathsf{RH} = \frac{\mathsf{p}_{w}}{\mathsf{p}_{s}}$$

$$p_w$$
 = partial pressure of water vapor
 p_s = saturated vapor pressure

The relative humidity of a particular gas sample will be sensitive to temperature and pressure changes. First of all, the partial pressure of the water vapor of a particular gas mixture is dependent on both temperature and pressure. From Dalton's Law and the Ideal Gas Law, we recognize that a change in total pressure of a gas sample effects a proportionate change in partial pressure of the water vapor. From the Ideal Gas Law, we see that a change in temperature changes the partial pressure of water in a closed system (the volume of the gas sample is constant). In an open system a change in temperature does not affect the partial pressure of the water vapor. Lastly, a change in temperature affects the value of the reference saturation vapor pressure used as the denominator of Equation 6.

Ambient relative humidity to treatment RH

When dry heat testing is done using a hot plate system or an air oven through which ambient air circulates, it is often desirable to know the relative humidity on the basis of the treatment temperature when we know the relative humidity of the atmosphere surrounding the hot plate or circulating through the oven. The equation relating relative humidity at ambient temperature, $RH_{(a)}$, to the relative humidity at test temperature, $RH_{(T)}$, is

$$\mathsf{RH}_{(\mathsf{T})} = \frac{\mathsf{RH}_{(a)} \, \mathsf{P}_{\mathsf{s}(a)}}{\mathsf{P}_{\mathsf{s}(\mathsf{T})}}$$
[7]

p_{s(a)} = saturation pressure at ambient temperature

This equation holds as long as the total pressure at the ambient condition and the total pressure at the test condition are equal. Garst and Lindell (5) have developed a chart for the conversion from relative humidity at one condition to relative humidity at another condition. Their chart is reproduced as Fig. 1.

When the ambient air is contained in a closed system as the gaseous environment to be heated to test temperature T without any other source of water, the relationship between ambient and treatment relative humidities will be

$$RH_{(T)} = RH_{(a)} \frac{P_{s(a)} - T}{P_{s(T)} - T_{a}}$$
[8]

T_a is the temperature of the ambient

Vapor density to relative humidity

From the Ideal Gas Law the partial pressure of water vapor behaves according to the equation

$$p_{w} = \frac{n_{w}}{V} \quad \text{RT}$$

Thus, percent relative humidity can be linearly related to a given concentration measure:

$$%RH_{(T)} = \frac{p_w}{p_{s(T)}} \times 100\% = \frac{n}{V} \frac{RT}{p_{s(T)}} 100\%$$
 [10]

or more generally

$$\% RH_{(T)} = C \frac{T}{P_{s(T)}} \delta$$
 [11]

C = concentration of water (units specified in Table 1)

- δ = constant of proportionality which depends on units of C (see Table 1)
- $p_{s(T)}$ = saturation pressure of water at temperature T in mm Hg
 - T = temperature in degrees Kelvin (°K)

Units of C	δ				
<u>moles</u>	62.37 <u>mm Hg</u> <u>liter</u>				
liter	℃ g-mole				
g	3.46 x 10 ³ ^{mm Hg} cm ³				
ml	⁰K g				
lb _m	9.59 × 10 ⁴ ^{mm Hg} ft ³				
ft ³	⁰K lb _m				

Specific humidity to relative humidity

The absolute humidity, w, can be calculated from

$$w = \frac{w_{\text{water}}}{w_{\text{air}}} = \frac{18.016 \text{ p}_{w}}{28.967(\text{P}-\text{p}_{w})}$$
[12]

wwater = weight of water vapor per unit volume

wair = weight of dry air per unit volume

pw = partial pressure of water vapor

P = total pressure

18.016 = molecular weight of water vapor

28.967 = molecular weight of dry air

If the water content is given in terms of absolute humidity, w, we can calculate $RH_{(T)}$ using the following equation:

$$RH_{(T)} = \frac{28.967 \text{ w P}}{p_{s(T)} \quad (18.016 + 28.967\text{w})}$$

$$w = \text{humidity as lb of water vapor per lb}$$
of dry air

Parts per million to relative humidity

An equation relating water content expressed as ppm of water to relative humidity at a temperature can be obtained by combining equations 4 and 6. The resulting equation is

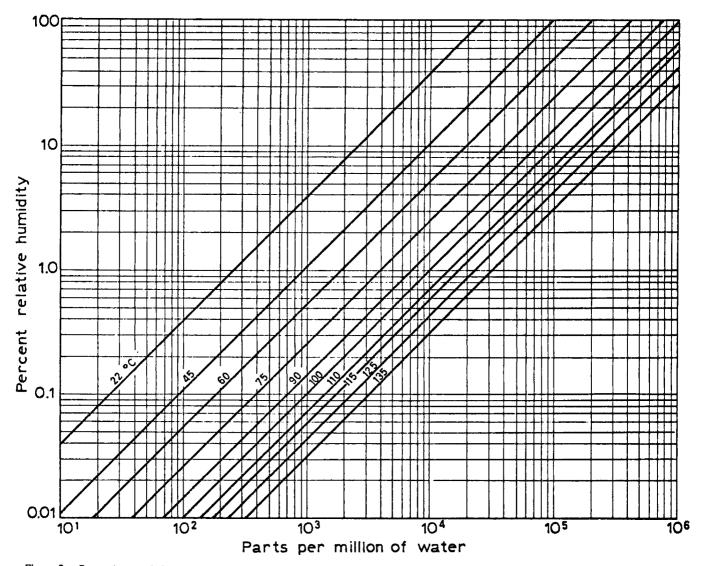


Figure 2. Conversion graph for parts per million of water and relative humidity for an open system (P = 760 mm Hg).

$$%RH_{(T)} = \frac{ppm \times P}{10^4 \times P_{s(T)}}$$
 [14]

For an open system where the total pressure is constant at one atmosphere, this relationship is linear and equation 14 becomes

[15]
%RH_(T) =
$$\beta_{(T)}$$
 ppm
e temperature dependency of $\beta_{(T)} = \frac{760}{10^4 P_{s(T)}}$

Where an atmosphere is 100% water vapor (10^6 ppm) and the temperature is above 100 C at one atmosphere of pressure (p = 760 mm Hg), a superheated water vapor condition exists. The relative humidity will be the total

th ca pressure (760 mm Hg) divided by the saturation pressure corresponding to the gas temperature. This condition is found in Figure 2 on the 10⁶ ppm water content line.

TABLE 2. Values of $\beta(T)$ at Various	Test	Temperatures
--	------	--------------

тс	P _{s(T)} [mm Hg]	β(T)
22	19.80	3.827 × 10 ⁻¹
45	71.83	1.058×10^{-1}
60	149.35	5.088 × 10-4
75	289.05	2.629 × 10 ⁻⁴
90	525.78	1.445 × 10-4
100	760.00	1.000 × 10-4
110	1074.56	7.072 × 10-5
115	1267.98	5.993 × 10-
125	1740.93	4.365 × 10-5
135	2347.26	3.237 × 10-

For a closed system that is sealed at ambient pressure and temperature (T_a) , the relationship at other temperatures (T) is more complicated since pressure will also change with temperature. The final total pressure will be the sum of the partial pressures of the component gases. For the dry gas component, the final partial

⁵The total pressure was taken as 760 mm Hg or I standard atmosphere. This total pressure varies depending on weather and location.

WATER CONTENT IN GASEOUS SYSTEMS

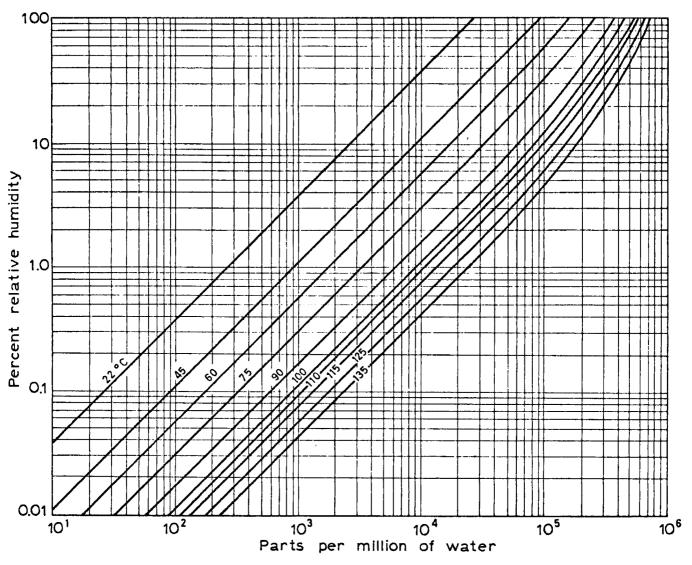


Figure 3. Conversion graph for parts per million of water and relative humidity for a closed system (p_{dg} = 760 mm Hg).

pressure, $p_{dg(T)}$, upon being heated, increases in accordance with

$$p_{dg(T)} = p_{dg(a)} \frac{T}{T_{(a)}}$$
[16]

p_{dg(a)} = the partial pressure of the dry gas at the time of sealing

This equation is only valid for closed systems where the gaseous volumes remain constant. By definition of relative humidity, the partial pressure of water at test temperature is

$$p_{w} = \frac{\% R H_{(T)}}{100} p_{s(T)}$$
[17]

Presumably, the relative humidity at test temperature $(RH\%_{(T)})$ is calculatable from the amount of liquid water added (see equation 11), and/or the relative humidity of the ambient gas (see equation 8) which is sealed in the closed system. At any rate, the total pressure will be

$$P = p_{dg(a)} \frac{T}{T_{(a)}} + \frac{\% RH_{(T)}}{100} p_{s(T)}$$
[18]

Example. Thus, for a closed system having been sealed at 22 C with the dry air having a partial pressure of 760 mm Hg, the total pressure at test temperature is

$$P = 760 \frac{T}{295} + \frac{\% RH_{(T)}}{100} p_{s(T)}$$
[19]

[20]

By combining equations 14, 17, and 19 the relationship necessary to convert from ppm to %RH is

$$%RH_{(T)} = \frac{\frac{T}{295} \beta(T)ppm}{1 - \frac{ppm}{10^6}}$$

Figures 2 and 3, respectively, show the relationship of water in ppm to %RH for an open and for a closed system. The results are linear for the open system and nonlinear for a closed system, especially at the larger values of ppm.

Dew point to relative humidity

Some researchers report water content in terms of dew point or the temperature at which condensation starts upon an isobaric cooling (cooling at constant total pressure) of the gas mixture. Since the pressure at dew point equals the partial pressure of water vapor, relative humidity can be calculated as

$$RH_{(T)} = \frac{P_{s} (DPT)}{P_{s}(T)}$$
[21]

Figure 1 shows the dew point temperature for gas mixtures of varying temperatures and relative humidities.

Psychrometric forms to relative humidity

Wet and dry bulb psychrometry forms are basic to most common humidity measurements. Conversion of wet and dry bulb temperatures to relative humidity relies heavily on use of psychrometric charts (2, 9). Such charts generally describe, for the user's information, the proper technique to determine relative humidity for any psychrometric determination.

SUMMARY

The common methods of measuring water in water-air systems are described. Equations are developed that

make possible the conversion of data from the different type of measuring unit into percent relative humidity. The location of the relevant conversions can be found by consulting Table 3. Equations to be used to carry out the measuring unit conversions are listed in Table 3.

TABLE 3.	Equations	to	be	used	to	convert	from	the	several
measuring u	nits to perce	ent i	relat	ive hu	mid	ity at trea	itment	temp	verature

Measurement	Equation number				
unit	In open system	In closed system			
Ambient RH	7	8			
Dew point	21	21			
Parts per million	15	20			
Specific humidity	13	13			
Vapor density	11	11			

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