ON THE NATURE AND SIZE OF PARTICLES IN HAZE, FOG, AND STRATUS OF THE LOS ANGELES REGION \mathbb{I}^2

M. NEIBURGER AND M. G. WURTELE

Depaitmed of *lIefeorology, Cnxeiszty* of *Calzfoinza ut Los Angeles, Los Angeles, Culifoinza*

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Measurements of relative humidity in fog and stratus cloud frequently give values far below 100 per cent, particularly in industrial areas. Drop radii measurements in the cloud show most frequent values around 7μ . The occurrence of drops in equilibrium at a relative humidity of 90 per cent would require solutions of concentrations of 17 per cent for sodium chloride and **23** per cent for sulfuric acid. Particularly in the latter case, i.e., for combustion nuclei, this high concentration seems unlikely. The hygroscopic nuclei would be about one-half the size of the drop.

Visibility measurements, in conjunction with the humidity, give a means of estimating the relative contribution of hygroscopic and non-hygroscopic particles in obstructing vision, on the assumption that the number of hygroscopic particles is constant. The observations indicate that the latter assumption is not valid. Using a relationship between liquid water content and visibility shown by Radford to fit observed data, a method is derived for determining the number and size of the hygroscopic particles, the value of the extinction coefficient due to non-hygroscopic particles, and the mass of the nucleus. Using the observations of humidity and visibility at Los Angeles Airport, and assuming that the nuclei are sodium chloride, these quantities are determined. The drop radii vary from 8 microns at 98 per cent humidity to 3 microns at' *6i.5* per cent, and the number decreases from **24** to *7* per cubic centimeter.

I. ISTRODUCTIOS

In the California Stratus Investigation of 1944 (11) it was noted that the maximum relative humidity recorded in the cloud at different stations in the vicinity of Los Angeles was frequently less than 100 per cent. While at Santa hlaria, a rural station, the recorded humidity reached 100 per cent in most of the soundings through stratus, there nere a number of soundings in u-hich it did not. $At U.C.L.A.,$ in the western part of Los Angeles, and in Pasadena, to the east of it, the highest humidity recorded in the cloud was most frequently about 90 per cent. It was suggested, without careful consideration, that "a plausihle explanation of this difference lies in the fact that there is little industrial activity in the vicinity of Santa Maria, but in the Los Angeles Basin there are a great number of factories injecting combustion products and other formi of hygroscopic nuclei into the air." When this explanation is examined, however, serious doubts are raised as to its validity, as we shall show below.

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The idea that combustion nuclei may lower considerably the humidity at which condensation occurs is presented quite generally in the literature. Thus Shaw (17) says, "It is hardly safe to assert that there is any limit of humidity below which clouds of water particles could not be formed in the air of industrial cities." On the other hand Kright (22) attempted to evaluate the contribution of sea-salt nuclei, combustion nuclei, and non-hygroscopic particles from average visibilities at coastal and inland stations in Britain in summer and winter, and concluded that "Kater fogs are due to sea-salt nuclei and if supersaturation is attained the fogs may thicken considerably owing to the deposition of water on the sea-salt nuclei which consequently become very much larger. Such deposition does not occur on the combustion nuclei if sea-salt nuclei are present."

11. OX THE XATURE AND SIZE OF SUCLEI

The three factors altering the equilibrium vapor pressure over drops from that over a plane pure water surface at the same temperature are curvature of surface, electric charge, and dissolved substances. Of these factors the first tends to raise the equilibrium vapor pressure, and the other two to reduce it.

The curvature effect is given by the formula

$$
\ln \frac{e_r}{E} = \frac{2\gamma}{\rho RTr} \tag{1}
$$

where e_r and E are, respectively, the equilibrium vapor pressures over a drop of radius *r* and a plane liquid surface at absolute temperature T , ρ is the density, γ is the surface tension of the liquid, and *R* is the gas constant. For pure water at 15° C. this formula becomes

$$
\ln \frac{e_r}{E} = \frac{1.11 \times 10^{-3}}{r}
$$

where r is expressed in microns. A 1-micron drop would thus have an equilibrium vapor pressure 1.001 times that for a plane water surface, i.e., would require a "supersaturation" of 100.1 per cent relative humidity in order to be in equilibrium. For larger drops the amount of supersaturation required is still less.

Figure 1 shows the frequency distribution of drop sizes measured by the sooted-slide method at several levels in a stratus cloud (12). The most frequent radius at all levels except near the cloud base is about 7 microns, and smaller drops were much less frequent. The preponderance of drops of one size is to be expected for drops formed by condensation, since larger drops will grow at the expense of smaller ones, other conditions being the same. The example shown in the figure is typical of the several cases in which measurements were made in California stratus. Thus for this cloud the drops are so large that the effect of curvature may be neglected.

That the effect of electric charge is also negligible may be seen by computing the charge required to offset the effect of curvature. If the drop has a charge of *q* electrostatic units, equation 1 becomes

$$
\rho RT \ln \frac{e_r}{E} = \frac{2\gamma}{r} - \frac{q^2}{8\pi r^4}
$$

To offset the curvature effect in a 1-micron drop would require 1.3×10^5 times the charge of one electron. For a 7-micron drop it would require 2.36×10^6 electronic charges. It is unlikely that such large charges ever occur. The effect of solutes (Raoult's law) is the only one remaining which might explain the occurrence of drops at less than 100 per cent relative humidity.

For solutions the reduction in rapor pressure is given by

$$
\frac{E}{e_s} = 1 - CM \tag{2}
$$

where e_s is the equilibrium vapor pressure for pure water, *M* is the molar concentration of the solution, and C is a factor which depends on the concentration,

FIG. 1. Drop-size distribution in one case of California stratus, as measured by the sooted-slide method.

temperature, and nature of the solute. $E/e_s = H$ is the relative humidity with respect to a plane pure water surface. If c is the mass concentration, $c = mM$, where *m* is the molecular weight. Then $H = 1 - cC/m$

$$
H = 1 - cC/m \tag{2'}
$$

The two types of solute which have been most frequently suggested as condensation nuclei under atmospheric conditions are sea salt, of which sodium chloride is the principal constituent, and combustion products, of which sulfuric acid is regarded as the most hygroscopic. For *M* of the order of 0.002 at **20°C.,** $C = 34$ for sodium chloride and $C = 43$ for sulfuric acid (6).

The concentration of solution in equilibrium at 90 per cent relative humidity is found by substituting 0.9 for E/e_s in equation 2. We get

$$
c = 0.1 \ m/C
$$

which for sodium chloride and sulfuric acid gives the concentration of **17** per cent and **23** per cent, respectively. These high concentrations seem surprising. If the fog drops consist of acid of this concentration, the corrosive effects would have been noticed. Since they have not, it is reasonable to reject the existence of such high concentration of acid.

If we characterize the size of the nucleus by the radius r_s of a sphere of equal mass, we can compute the ratio of nucleus to drop size which will result in this concentration. The mass of the solute in one drop is given by the two expressions

$$
m_s = \frac{4}{3}\pi r^3 \rho c = \frac{4}{3}\pi r_s^3 \rho_s
$$

Thus

$$
\frac{r_s}{r} = \sqrt[3]{\frac{\rho C}{\rho_s}}
$$
 (3)

The following table gives the values of this ratio for sodium chloride and sulfuric acid for the concentrations found above, and the corresponding nuclear size and mass for 7-micron drops.

Since John Aitken's invention of the dust counter there have been hundreds of studies of the properties of "nuclei" (for summaries see Landsberg (9) and Simpson (18)). Most of these, however, have been concerned with those whose size can be determined from their ionic mobility, i.e., of radius of the order 10⁻⁶ cm. or less (see, for example, Israel and Schulz (7) or Nolan and Guerrini **(13)).** The properties of larger nuclei are, paradoxically, more diffcult to determine and require more circuitous methods. Kohler **(8),** by measurements of corona and chemical analysis of frost deposits obtained when supercooled fog passed over objects, concluded that the nuclei were particles of sea salt with "radius" of the order 10^{-5} cm. Similar results were obtained by Wright $(21, 21)$ **22, 23)** by an analysis of observations of humidity and visibility. Simpson **(19)** raised objections to Wright's results. With respect to Kohler's measurements, there is considerable question of the applicability of the formula he used relating corona diameter and drop size to the range of sizes involved. In table 1 are presented the ranges of values reported by these and other observers, and now tentatively accepted.

The measurements of nuclei thus have values considerably smaller than those necessary to maintain equilibrium of drops of solution at 90 per cent relative humidity. In particular, the combustion nuclei have been found to be at largest less than $\frac{1}{30}$ the size required. Nuclei of sea salt have been found to be quite a bit larger than combustion nuclei. We must conclude that combustion nuclei cannot be responsible for the difference in humidity measured in the stratus at Los Angeles and Santa Maria.

The possibility that the low humidity measurements in the Los Angeles area are due to instrumental errors was considered. It was pointed out in the 1944 report that this is rendered doubtful by the fact that the same types of instru-

* The curious concentration at the humidities **07** per cent and 94 per cent in columns two and four may be due to the fact that they may be read from the psychrometric tables without interpolation.

ments and methods of evaluation were used at all stations. Besides this, surface observations show the existence of fog at humidities considerably below 100 per cent, even at rural stations, with more frequent low humidities in fog in industrial areas. Thus Pick (15, 16) showed that 20 per cent of all thick fogs observed at 0'700 at Cardigan, England, in 1929-30 mere accompanied by

relative humidities of **90** per cent to **93** per cent, and that the majority of all fogs occurred in unsaturated air. He also quoted observations of thick and dense fogs at sea with humidities in the *80's.*

That the difference between purely coastal and industrial areas exists with respect to surface observations of fog and humidity in the Los Angeles region is shown by comparison of stations relatively free from industrial influence with those so affected. The two groups of stations considered were CIT, Ontario, and San Bernardino as inland locations and Los Angeles Municipal Airport, Oceanside, and Santa Barbara on the coast. Table **2** lists the frequency of observed (wet-and-dry-bulb psychrometer) relative humidities at the time of lowest visibility on each morning during a period of several months, provided this visibility was 1 mile or less.

REPORTED VISIBILITY	NUMBER OF OBSERVA- TIONS AT INLAND STATIONS	PER CENT OF TOTAL	NUMBER OF OBSERVA- TIONS AT MARINE STATIONS	PER CENT OF TOTAL
miles				
0	5	8.6	16	20.8
1/8	8	13.8		9.1
1/5	$\overline{2}$	84.4 3.4	$\overline{2}$	41.6 2.6
1/4	5	8.6	7	9.1
1/2	12	20.7	12	15.6
3/4	10	17.3 65.6	14	$18.2 \, 58.4$
	16	27.6	19	24.6
$Total \dots \dots$	58	100.0	77	100.0
Average visi- bility	$9/16$ miles		$7/16$ miles	

TABLE *3 Frequency* of *minimum visibilities reported for data in table 8*

This distribution must be compared with a similar one for the visibilities, with intervals determined by standard airway observation practice, shown in table **3.**

While the visibilities at marine stations were somewhat lower than at inland stations for the cases considered, the difference is not nearly so marked as that in the humidities. We see that while only **19.5** per cent of the cases of low visibility had humidities below 95 per cent at marine stations, the corresponding value was **58.6** per cent at inland stations.

With the hygroscopic action of combustion nuclei rejected as a factor in explaining the more frequent existence of cloud drops at low humidities, and further evidence that the phenomenon is real, me are led to seek other explanations. One hypothesis which might be the explanation is that the drops are not in equilibrium, but have not had time to evaporate completely at the time of observation. Findeisen (1, **2)** derived an equation for the time of evaporation of drops. For relative humidity **90** per cent, temperature 5"C., and pressure

900 mb., his formula is

$$
t\,=\,0.053r^2
$$

where *r* is in microns and *t* in seconds. Seven-micron drops would thus take *2.5* see. to evaporate under these conditions, and from this Findeisen concludes that "the probability of small drops being present in air only slightly below saturation is practically zero."

111. VISIBILITY **.4ND** PARTICLE **SIZE**

The preceding material points up the desirability of direct measurement of the nature and size of condensation nuclei. As was pointed out above, such direct measurement is extremely difficult. However, Wright's approach may be applied to the determination of the drop and nucleus size, if assumptions are made as to the nature and number of active hygroscopic particles. In this may hypotheses regarding their nature and number can be tested, using only the regular Weather Bureau observations of visibility and humidity.

The relationship between the visibility (visual range) and the suspended particles which limit it is given by Koschmieder's formula (10)

$$
V = \frac{1}{\sigma} \ln \frac{1}{\epsilon} = \frac{3.91}{\sigma} \tag{4}
$$

where *V* is the visibility in centimeters, σ is the extinction coefficient due to suspended matter, and ϵ , the threshold of perceptible contrast between the apparent brightness of an object and the brightness of the background, is taken to be 0.02. The extinction coefficient σ is contributed to by (1) scattering by molecules, *(2)* scattering and absorption by hygroscopic particles and their associated water drops, and (3) scattering and absorption by non-hygroscopic particles of dust, smoke, etc. We may write

$$
\sigma = \sigma_M + \sigma_N + \sigma_P
$$

where σ_M , σ_N , and σ_P are the separate extinction coefficients for processes (1), *(W),* and *(S),* respectively.

For average wave length of light and normal temperature and pressure Hulburt (5) computed $\sigma_M = 1.6 \times 10^{-7}$ cm.⁻¹ For water drops σ_N is given by Stratton and Houghton *(20)* to be (see appendix)

$$
\sigma_N = 2\pi Nkr^2 \tag{5}
$$

where *N* is the number of drops per cubic centimeter and πr^2 the area of each, and *k* is a factor depending on the ratio of *r* to the wave length of the light. For *r* several times the wave length, *k* departs only slightly from **1. A** similar expression would hold for drops consisting of solutions, but with a different value of *k*, and presumably also for σ_P , with a still different factor. We shall assume that in both these cases *k* is also near unity for the wave lengths in which we are interested.

Equation **4** may now be written

$$
\sigma_{\rm P} + 2\pi N r^2 = \frac{3.91}{V} - 1.6 \times 10^{-7}
$$
 (6)

For values of visibility up to *25* km. the second term on the right is negligible. For convenience we shall absorb it in σ_P . The visibility is observed, and the unknowns in this equation are σ_P , N, and r. If successive measurements are made in the same air at different humidities, but away from sources of pollution, *r* and *N* may change, but σ_P should remain constant. The change in drop radius will be in accord with equation 2'. Since the concentration in the drop is given by

$$
c = 3m_s/4\pi r^3 \rho
$$

we obtain from equation **2'**

$$
r = K[m_s/(1-H)]^{1/3}
$$
 (7)

where $K = (3C/4\pi\rho m)^{1/3}$ is a parameter of the nuclear substance which has a variation of the order of 1 per cent with temperature and concentration. For sodium chloride and sulfuric acid it has the values *0.50* and *0.45,* respectively.

Substituting for *r* from equation **7** in equation 6 we obtain

$$
\sigma_P + 2\pi N K^2 [m_s/(1-H)]^{2/3} = 3.91/V \tag{8}
$$

If m_s and N are constants with changing humidity, as well as σ_P , the equation gives the relationship between humidity and visibility in the form

$$
(1 - H)^{-2/3} = AV^{-1} + B
$$

where *A* is a parameter depending on *N*, *K*, and m_s , and *B* depends on σ_P . If a value of σ_P is assumed, the curve representing this equation is determined by one observation; other observations may be used to check it. The value $\sigma_P = 0$ leads to the familiar curves of Wright (figure **2).**

For the purpose of testing this relationship hourly observations of visibility and humidity at the Los Angeles Municipal Airport were tabulated. Only days on which there mere periods with visibilities *2* miles or less were used, and precautions were taken to eliminate effects which would alter σ_P . For instance, if a sea breeze brought about a change of air mass over the station, or if advection of smoke was evident, the observations were not used. Thus σ_P might change from day to day, but not during the series of hours in which the variation of visibility with humidity on a single day was tabulated, insofar as it was possible to select data so as to avoid it. The visibilities were grouped and averaged by *5* per cent intervals of humidity. The results are shown by the broken line in figure 3.

Using the point on the curve for 85 per cent relative humidity to determine Λ , the theoretical curves representing equation **8** were computed and drawn for two values of σ_P . It is seen that while these curves, particularly the one for $\sigma_P = 2 \times 10^{-7}$, agree fairly well with the observations for high humidities, they

diverge widely from them for lower humidity values. To get a closer fit it would be necessary to assume σ_P negative, a physically unacceptable assumption.

Thus the hypotheses of σ_P and N constant must be reconsidered. Since the data were selected with an attempt to keep σ_P constant, and since the constant N requires σ_P to be negative, it is logical to abandon the second hypothesis first. KO alternative suggestion has been made regarding the variation of *N,* nor does one arise from physical reasoning. However, W. H. Radford's compilation of measurements of the content of liquid water in fogs (4) gives a clue to the behavior of drops under varying humidities. He plotted observations of visibility against observations of liquid content, both on a logarithmic scale, and found the

FIG. 2. Relation between relative humidity and visibility, assuming N constant and $\sigma_p = 0$ (after Wright).

curve of best fit to be a straight line (figure 4). Thus if w is the liquid content in grams per cubic centimeter of air, there is empirical evidence that $\ln V = \ln \frac{g}{g}$ $a + b$ ln w or, more conveniently,

$$
V = awb \t\t(9)
$$

Radford does not give the constants *a* and *b,* but from his graph it is found that *^a*= **0.032** and *b* = **1.43.**

This empirical relationship may be incorporated into our system of equations in place of the assumption $N = constant$. The liquid content is

$$
w = \frac{4}{3}\pi \rho r^3 N \tag{10}
$$

and if *N* is eliminated between this equation and equation 6 we have

$$
\sigma_P + \frac{3w}{2\rho r} = \frac{3.91}{V} \tag{11}
$$

or, from equation 9

$$
\sigma_P + \frac{3}{2\rho r} \left(\frac{V}{a}\right)^{1/b} = \frac{3.91}{V}
$$

FIG. **3.** Observed and computed relations between relative humidity and visibility

Using equation **7,** we may write

$$
\frac{r_1}{r} = \left(\frac{1-H}{1-H_1}\right)^{1/3} \tag{12}
$$

where r_1 is the radius at humidity H_1 , and r that at H . Thus our equation becomes

$$
\sigma_P + \frac{3}{2\rho r_1} \left(\frac{V}{a}\right)^{1/b} \left(\frac{1-H}{1-H_1}\right)^{1/3} = \frac{3.91}{V} \tag{13}
$$

This gives the relation between humidity and visibility on the basis of our new assumption, giving an implicit relationship between V and H , with σ_P ,

 r_1 , and H_1 as parameters. By choosing two points on the observed curve, H_1 and H_2 , and using the observed visibilities V_1 and V_2 , the remaining quantities σ_P and r_1 can be determined. By choosing 85 per cent and 67.5 per cent, it was found that $\sigma_P = 0.14 \times 10^{-5}$ cm.⁻¹, or 0.14 km.⁻¹, and $r_1 = 4.08 \times 10^{-4}$ cm. = 4.08μ . The curve representing the equation with these values of the parameters

FIG. **4.** Relation between liquid water content and visibility, according to summary of **observations** by Radford.

is shown in figure **3** also. It comes very close to the observed values, not only at the two points for which it was fitted but in its entire range.

Using r_1 as determined from equation 13 the values of r may be evaluated from equation 12. The mass of solute m_s is determined from equation 7. The value of *K* for sodium chloride was used in this connection, and whenever else the solute parameters enter. To evaluate N , we use equations 9 and 10, inserting the values of *r* computed for the observed values *V.* The values of these quantities at the different humidities are presented in table 4, along with the computed and observed visibilities.

It is interesting to note that the drop radius determined in this way for fog at

98 per cent relative humidity, 8μ , is almost exactly the modal radius found for drops in stratus by direct sampling. Even at relatively low humidities the drops remain as large as **3** microns, and in fact the "radius" of the solid nucleus is computed to be **2** microns.

The decrease of the number of drops from **23.5** per cubic centimeter at 98 per cent to **7** per cubic centimeter at 67.5 per cent is not readily explained physically. This, too, corresponds to a phenomenon in the stratus observations. In those observations it was found that while the drop size mas constant with height, the liquid content increased linearly from base to top. This means that the number of drops must be greater at the top of the cloud than the bottom. The

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Mean values of *various quantities pertaining to suspensoids in the atmosphere (Los Angeles Municipal Airport)*

 σ_P = extinction coefficient due to non-hy- $r =$ radius of hygroscopic particles. groscopic particles. $m_{\lambda} = \text{mass of solute.}$

temperature is lower and humidity presumably higher at the top than the bottom.

The value of σ_P , 0.14 km.⁻¹, is a reasonable one, corresponding to a visibility of 16 miles in the absence of the larger hygroscopic particles. When the marine air containing these large particles is entirely absent in Los Angeles, the visibility is upward of **30** miles in the absence of concentration of industrial pollutants as well. It is seen that in cases of sea haze and fog, if this evaluation is correct, non-hygroscopic particles contribute at most one-fourth to the opacity of the air, and at higher humidities, an insignificant amount. Nothing can be said as to size and number of these particles; if there are 100 per cubic centimeter, the *r* would have to be about 0.4 micron, while 1000 per cubic centimeter would require, for this value of $\sigma_{\mathbf{F}}$, about 0.1 micron radius. It is planned to conduct direct counts to provide additional information regarding these quantities.

In figure **3** it will be seen that the visibility increases rapidly as the humidity decreases to **67.6** per cent, and there becomes approximately constant. This might be considered the value at which the drop becomes crystalline, and thus would give (within *5* per cent, because of the grouping of data) the humidity in equilibrium with a saturated solution of the solute. If the solute mere pure sodium chloride, this humidity should be *76* per cent. The experimental determination of the humidity over a saturated solution of sea salt has been carried out by $J. S.$ Owens (14) . The experiment was carried out several times at varying temperatures, with the results listed in table *5.* Owens' values are thus in complete agreement with the data of figure **3.**

IV. CONCLUSION

The methods employed have indicated that it is not combustion nuclei acting alone vhich produce the difference between humidities in fog and stratus in

TABLE a						
	Relative humidities over saturated solutions of sea salt (after J. S. Owens)					
RANGE OF TEMPERATURES	RANGE OF HUMIDITIES	MEAN HUMIDITY				

T-IBLE *5*

OF. **per** *cent per* cent $53-66$ 60-71 67

industrial and rural districts. Large nuclei of sea salt would explain the observed variation of visibility with humidity but cannot explain the difference between rural and industrial areas. The hypothesis that the same number of hygroscopic nuclei are active at all reasonably high humidities must be rejected in favor of one which states that the number increases rather rapidly with humidity. In this connection it should be noted that the methods used did not take account of the contribution of the hygroscopic particles which become inactive to the opacity.

If the methods are valid, the data indicate that the drops in fog at the Los Angeles Municipal Airport are of the same size as those found in stratus over the ocean in this region, and in measurements in fog and stratus elsewhere. This would suggest that there is a preferred size for drops in stable air clouds, and an increase in condensation due to further cooling results in more nuclei becoming active, rather than the further growth of existing drops.

Finally it should be emphasized that the numerical results are significant only in order of magnitude, as is shown in the appendix. If greater accuracy is to be attained by the methods employed in this paper, it would seem that further study of drop-size distributions is required. Such a study would necessarily involve consideration of the phenomena discussed in the tvo preceding paragraphs.

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APPENDIX

The assumption made at this point, usually tacit **in** the literature, is that the proper value of the total surface area of n drops,

$$
\pi(r_1^2 + r_2^2 + \cdots r_n^2) = \pi \sum_{1}^{n} r_i^2
$$
 (14)

can be adequately approximated by $\pi n \bar{r}^2$, where \bar{r} is the arithmetic mean of the n radii. The algebraic identity relating these quantities is

$$
n\bar{r}^2 - \sum_{i=1}^{n} r_i^2 = - \sum_{i=1}^{n} (r_i - \bar{r})^2
$$

so that if there is any dispersion at all there will be an error, which will always have the same sign (negative), with a magnitude equal to *n* times the variance of the distribution. Calculations of this magnitude from empirical distributions of direct measurements recorded by two observers are presented in column **3** of table **6.** It turns out to be rather large, in one instance amounting to **32** per cent.

However, the expression 14 does not enter explicitly into equation 11; rather the substitution from equation 10 results in a different approximation:

$$
\sigma_N \propto \sum r_i^2, \qquad w \propto \sum r_i^3
$$

Thus

$$
\sigma_N = w \, \frac{\sigma_N}{w} \propto w \, \frac{\sum r_i^2}{\sum r_i^3}
$$

This quantity has been approximated by w/\bar{r} . The percentage error is therefore

$$
1-\frac{\bar{r}\sum r_i^2}{\sum r_i^3}
$$

Empirically determined values for this error have been tabulated in column **4** of table **6.** Again it is large, but does not invalidate the order of magnitude of the results of the paper.