

Equation for Predicting the pH Value of Rainwater using Air Quality Index

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ABSTRACT: This research derives and validates an equation to predict rainwater pH using the Air Quality Index (AQI) based on Fick's law of diffusion. Considering the surface area and diffusivity coefficient parameters, the mathematical model equation improves the understanding of mass-transfer mechanisms between pollutants and rain droplets. Validating the results obtained in this work with existing data showed an initial prediction deviation of 47.3 %, which was further reduced to 41.06 % using a conversion factor. This result demonstrates the accuracy of the mathematical modeling reported in this work. The modified equation also offers a practical tool for environmental monitoring, aiding in the mitigation of acid rain effects. This work supports making decisions for environmental management and contributing significantly to atmospheric science by providing a quantitative method for assessing acid rain potential and guiding effective mitigation strategies.

KEYWORDS: Air Quality Index, Acid rain, pH prediction, Environmental monitoring, Atmospheric pollutants

<https://doi.org/10.29294/IJASE.10.4.2024.3722-3729> ©2024 Mahendrapublications.com, All rights reserved

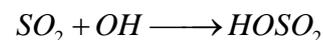
1. INTRODUCTION

Acid rain has been observed to cause widespread acidification [1], which poses a serious threat to various ecosystems [2]. In particular, the acid rain deposition on soil leads to increased soil acidity, negatively impacting soil microflora populations [3] and causing alterations in cation exchange dynamics under the influence of hydrogen ions. Moreover, acid rain poses a significant threat to structures made of marble, limestone, concrete, and metals like iron, copper, and zinc, as it accelerates corrosion processes [4]. Iconic landmarks such as the Taj Mahal in Agra and the Giant Buddha statue in China have suffered visible damage as a result of acid rain [5].

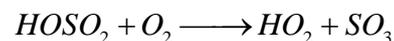
Exposure to these acids can cause various health issues, including irritation to the eyes, skin, and mucous membranes, as well as allergic reactions. Prolonged exposure may result in conditions such as delayed pulmonary edema, pneumonitis, and bronchitis [6-8]. While carbonic acid itself is not considered harmful to humans, its presence in the atmosphere is significant due to its role in the corrosion of metallic structures and its association with high

levels of carbon dioxide, a major factor in carbonic acid formation. During rainfall, the interaction between air pollutants and rainwater leads to the formation of acid rain that mainly contains the primary contributors to acid rain such as sulphuric acid, nitric acid and carbonic acid [3]. The chemical reactions for the formation of these acids are given below.

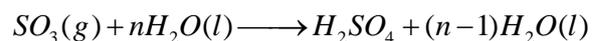
Formation of sulphuric acid:



which is followed by,



Sulphur trioxide (SO_3) is rapidly converted to sulfuric acid in the presence of water.



Formation of nitric acid:

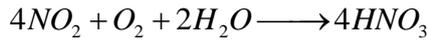
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Received: 10.04.2024

Accepted: 28.05.2024

Published on: 24.06.2024

Vinothkumar et al.,



Formation of carbonic acid:



Numerous studies have explored acid rain's causes, effects, controlling strategies, and case studies across various locations and periods [9-13]. But there is lack of efficient methods to predict the pH of acid rain to mitigate the effects on human beings. Hence this work aims to derive an equation using Air Quality Index (AQI) data to predict rainwater pH. This involves certain assumptions, discussed later, to enhance understanding of pollutant mass transfer into rainwater and provide a quantitative prediction method. Using the AQI for predicting rainwater pH offers several advantages by directly linking atmospheric pollution to rainwater chemistry. This predictive equation provides a practical and efficient tool for assessing acid rain potential, aiding researchers, environmentalists, and policymakers in anticipating and mitigating acid rain effects. Utilizing readily available air quality data, this approach can support early warning systems and targeted mitigation strategies. However, certain assumptions simplify the complex atmospheric interactions. This study mainly aims to derive an equation for predicting rainwater pH using AQI pollutant concentrations and bridges the gap between air pollution monitoring and rainwater acidity.

1.1. ASSUMPTIONS

- i. All the falling rain droplets are having uniform terminal velocities and same in size.
- ii. The mass transfer (diffusion) occurs only from air to water droplet and there is no transfer of mass occurs through diffusion from liquid to air.
- iii. The size of the water droplet and velocity remain constant until it reaches the ground.
- iv. The concentration of pollutants (C_i) in the rain droplet is not uniform, the concentration in point 1 (P_1) is higher and there is no concentration of pollutants at point 2 (P_2) as shown in Fig. 1.
- v. The concentration of air and amount of rain droplets is uniform over the entire atmosphere where the rainfall occurs.

vi. All the pollutants diffused are completely reacted with rain droplets.

2. METHODOLOGY

2.1 MEASURING THE SIZE OF THE RAIN DROPLET

Raindrop parameters such as shape, size, diameter, height, volume, surface area, thickness, and terminal velocity are critical for calculating mass transfer rates. Measurement techniques can be manual or automated. Manual methods involve direct observation, including photographic, oil immersion, flour pellet, and stain methods. Automated methods, such as optical scattering, imaging, and disdrometers, utilize advanced technologies for accurate measurements [14]. The Global Precipitation Measurement (GPM) mission by NASA and JAXA studies raindrop characteristics within clouds to gather predictive data before rainfall. GPM provides information on rainfall, snowfall, raindrop size distribution, and mass-weighted mean diameter [15]. GPM data is accessible to researchers, forecasters, and students, maximizing its utilization in research and applications [16]. Raindrop sizes typically range from 0.1mm to 6mm, with an average size of 3.05mm. Larger raindrops tend to break into smaller pieces [17-19] and this average size is often represented by the value of 3.05 mm in Fig. 1.

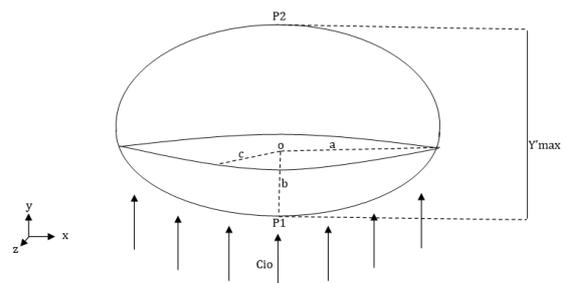


Fig 1: Shape of falling rain drop (3.05mm) from video disdrometer [19]

2.2 SURFACE AREA OF MOLAR FLUX

The surface area of the spheroid can be calculated using Knud Thompsen's formula [20],

$$surface\ area = 4\pi \frac{(a^p b^p + a^p c^p + b^p c^p)^{\frac{1}{p}}}{3} \quad 1$$

- $p \approx 1.6075$, (p has an error of 1.061%).

- a and b are the radius, c is the distance from the center of the rain droplet to point 1.

The mass transfer occurs in the bottom side of the rain droplet, the bottom side is in the shape of a semi-spheroid. For semi-spheroid the surface area formula changes as;

$$surface\ area = 2\pi \frac{(a^p b^p + a^p c^p + b^p c^p)^{\frac{1}{p}}}{3} \quad 2$$

p ≈ 1.6075

2.3 CONCENTRATION OF POLLUTANT USING AQI

In this work, Fick's law of diffusion is highlighted as a fundamental principle governing the rate of diffusion of substances across a given area. As stated in the law, it is crucial to determine the concentration of pollutants in order to determine the rate of mass transfer. The concentration can be quantified by using the air quality index measuring equation [21],

$$C_p = (I_p - I_{Lo}) \left(\frac{BP_{Hi} - BP_{Lo}}{I_{Hi} - I_{Lo}} \right) + BP_{Lo} \quad 3$$

2.4 DIFFUSIVITY MEASUREMENT

The diffusion coefficient is crucial for determining the mass transfer rate in gas-liquid systems, quantifying how quickly a substance diffuses from high to low concentration. Various experimental methods are used to accurately determine the diffusion coefficient of a gas into a liquid, involving controlled conditions and monitoring gas concentration changes over time to derive the coefficient for specific gas-liquid systems [22,23]. The diffusivity of major pollutants causing acid rain is listed in Table. 1.

2.5 DERIVATION

In this section, we derive an expression to predict the pH of raindrops formed from pollutant diffusion in the atmosphere [24]. The derivation is based on Fick's law, which describes the diffusion of substances in a medium. By applying certain assumptions and boundary conditions, we arrive at an equation that relates the pH of raindrops to the

concentration of pollutants and the terminal velocity of raindrops.

$$N_i = (N_i + N_R) x_i - D_{iR} \frac{dC_i}{dY} \quad 4$$

According to Fick's law as given in equation 4, the molar flux N_i of a binary mixture concerning a stationary observer is expressed as the difference between the convective flux ($N_i \times x_i$) and the diffusive flux ($-D_{iR} \times dC_i/dY$). This equation characterizes the net movement of pollutant molecules with respect to height (Y) in the atmosphere.

According to assumption-(ii) there is no mass transfer from rain droplets to air. Hence, the molar flux of raindrop N_R becomes zero. Hence the equation 4 becomes [$N_R = 0$]

$$N_i = N_i x_i - D_{iR} \frac{dC_i}{dY} \quad 5$$

We further simplify the equation 5 by expressing $N_i \times x_i$ as $C_i \times v_{ter}$ [25, 26], where C_i is the molarity of pollutant i and v_{ter} is the terminal velocity of raindrops:

$$N_i = C_i v_{ter} - D_{iR} \frac{dC_i}{dY} \quad 6$$

As per assumption (iv) there is no concentration of pollutants in the height of Y'_{max} (P2). This assumption is taken into consideration to break down the complexity. Integrating equation 6 by applying the below boundary condition [27,28];

- i. $C_i = C_{io}$ at $Y = 0$
- ii. $C_i = 0$ at $Y = Y'_{max}$

$$\frac{1}{D_{iR}} \int_0^{Y'_{max}} dY = \int_{C_{io}}^0 \frac{dC_i}{N_i - C_i v_{ter}} \quad 7$$

On further simplifying equation, we arrive

$$N_i = \frac{C_{io} \cdot v_{ter}}{1 - e^{-(Y'_{max} \times v_{ter} / D_{iR})}} \quad 8$$

The rate of mass transfer can be calculated as the product of the molar flux and the total surface area

$$M_i = S_{avg} \cdot N_i \quad 9$$

Substituting equation 8 in equation 9,

$$M_i = S_{avg} \cdot \left[\frac{C_{i0} \cdot v_{ter}}{1 - e^{-\left(\frac{Y'_{max} \cdot v_{ter}}{D_{iR}}\right)}} \right] \quad 10$$

To calculate the total moles of pollutants transferred during a raindrop's descent, we multiply the rate of mass transfer by the descent time. This product represents the cumulative pollutant transfer, giving the total moles reaching the ground with the raindrop [2].

$$W_i = S_{avg} \cdot \left[\frac{C_{i0} \cdot v_{ter}}{1 - e^{-\left(\frac{Y'_{max} \cdot v_{ter}}{D_{iR}}\right)}} \right] \cdot t_g \quad 11$$

For total amount of pollutant,

$$W = \sum_{j=i} W_i = \sum_{j=i} S_{avg} \cdot \left[\frac{C_{i0} \cdot v_{ter}}{1 - e^{-\left(\frac{Y'_{max} \cdot v_{ter}}{D_{iR}}\right)}} \right] \cdot t_g \quad 12$$

This research aims to determine the pH value of raindrops, reflecting their acidity or basicity due to atmospheric pollutants. Pollutants (SO_2 , NO_2 , CO_2) react stoichiometrically with water to form equal amounts of acids (H_2SO_4 , HNO_3 , H_2CO_3). Assuming complete reaction, total acid formation is expressed accordingly. To find the pH value, we calculate the molarity of pollutants in the raindrop by dividing the total moles of pollutants by the raindrop's volume.

Molarity = Total moles of pollutant / volume of rain drop 13

$$molarity = \frac{\sum_{j=i} W_i}{V_R} \quad 14$$

By finding the molarity we are able to find the amount of hydrogen ions are dissociated when the acid is incorporated with water. pH denotes the potential of hydrogen.

$$pH = -\log_{10} [H^+] = -\log_{10} [molarity] \quad 15$$

Knowing the molarity and total moles of pollutants doesn't accurately determine pH due to the non-proportional relationship. We introduce the parameter "n" to distinguish between monoprotic and diprotic acids. Monoprotic acids have acidity directly proportional to the moles, while diprotic acids

release twice as many hydrogen ions (H^+), doubling the acidity. Sulphuric and carbonic acids are diprotic ($n=2$), and nitric acid is monoprotic ($n=1$) [26-28]. By incorporating "n" into our calculations, we accurately account for this distinction and calculate pH based on the molarity and unique characteristics of these acids.

$$pH = -\log_{10} [n_i \cdot molarity] \quad 16$$

where,

$n_i = 1$, for monoprotic acids (nitric acid)

$n_i = 2$, for diprotic acids (sulphuric acid, carbonic acid)

substituting equations 12 and 14 in equation 16, we get,

$$pH = -\log_{10} \left[\frac{S_{avg} \cdot v_{ter} \cdot t_g \cdot \left(\sum_{j=i} \frac{n_i \cdot C_{i0}}{1 - e^{-\left(\frac{Y'_{max} \cdot v_{ter}}{D_{iR}}\right)}} \right)}{V_R} \right] \quad 17$$

[: $v_{ter} \cdot t_g = h_R$]

$$pH = -\log_{10} \left[\frac{S_{avg} \cdot h_R \cdot \left(\sum_{j=i} \frac{n_i \cdot C_{i0}}{1 - e^{-\left(\frac{Y'_{max} \cdot v_{ter}}{D_{iR}}\right)}} \right)}{V_R} \right] \quad 18$$

The above expression is the required equation for the prediction process. Where the parameters denoted with suffix "i" represent the individual pollutants. n_i is the parameter that represent the characteristics of acid formed by the diffusion of pollutants, whether the acid is monoprotic or diprotic.

3. RESULTS AND DISCUSSIONS

In our analysis, we emphasize the importance of utilizing equation 18 for pH calculation, which significantly helps to enhance environmental understanding and management decisions. To validate its efficacy, we analyzed the data reported by Shiba et al. [29]. The critical parameters essential for determining the pH value as mandated by equation 18 were substituted and the resulting pH values were compared.

Instead of using real-time rainfall data to verify the equation's accuracy, we relied on pre-existing data discussed in the report [29]. They have collected extensive data on the pH levels at various heights during rainfall and utilized a mathematical model of the raindrop acidification process based on the mass conservation of chemical species in raindrops. Hence, we compared this data with the pH values calculated using our derived equation. This approach ensures a robust validation of our model, leveraging established data to demonstrate the equation's reliability and practical utility in predicting rainwater acidity.

This dataset provided crucial parameters such as initial raindrop height, pollutant concentrations (SO_2 and HNO_3), raindrop size, and corresponding pH values. By incorporating these parameters into our equation and the data that have been used for the calculation are shown in Table. 2. Further, detailed comparative analyses have been conducted between calculated and observed pH values. The results of validation process ensure the reliability and practical utility of our equation in predicting rainwater acidity, thereby facilitating more effective environmental monitoring and mitigation efforts.

The critical parameters essential for determining the pH value, as mandated by equation 19, are substituted;

$$pH = -\log_{10} \left[\frac{1.004 \times 10^{-6} \times 1000 \times (8.16 \times 10^{-7} + 4.078 \times 10^{-9})}{2.679 \times 10^{-7}} \right] \quad 19$$

$$pH = -\log_{10}[0.0031] = 2.5124 \quad 20$$

The calculated pH value of 2.5124 significantly deviates from the target pH of 4.772 by 47.3 %, indicating a need for refinement. This discrepancy arises from the parameter n_i , representing the nature of acids (1 for monoprotic, 2 for diprotic), and the assumption of complete pollutant reaction with rainwater.

To address this, n_i was replaced with a conversion factor X . Unlike the discrete n_i , X offers a continuous depiction of pollutant conversion to acidic species, considering the stoichiometry of chemical reactions. This approach improves model accuracy and better predicts rainwater acidity based on air quality indices. Determining X requires understanding

the reaction rate and final pollutant concentration in raindrops. Focusing on the forward reaction pathway for clarity, we acknowledge potential oversights in pollutant dynamics. This simplified model aims to validate the derived equation, with room for improved pH predictions through accurate conversion factors.

Here the conversion X is found using the stoichiometry equation for the conversion of SO_2 and HNO_3 . The rate equation for the reactions is as follow,

$$-r_{so_2} = K_1 C_{SO_2} - K_1 \cdot C_{HSO_3^-} \cdot C_{H^+} \quad 21$$

$$-r_{HSO_3^-} = K_2 C_{HSO_3^-} - K_2 \cdot C_{SO_3^{2-}} \cdot C_{H^+} \quad 22$$

$$-r_{HNO_3} = K_3 C_{so_2} - K_3 \cdot C_{NO_3^-} \cdot C_{H^+} \cdot C_{H_2O} \quad 23$$

K_1 , K_2 and K_3 are the rate constant which has a value of 1.74×10^{-2} M, 6.24×10^{-8} M and 15.4 M [23]. Initially, there is no concentration of HSO_3^- , SO_3^{2-} , H^+ and NO_3^- in the rain drop so their concentration become zero in the rate equation, after the integration. The calculation process follows a systematic order to determine the final concentrations and conversions of SO_2 , HSO_3^- and HNO_3 . First, the known initial concentrations of SO_2 , HSO_3^- , and HNO_3 are substituted into the rate equations along with their rate constants and the time taken for raindrops to reach the ground. The conversion of SO_2 is computed first, impacting the concentration of HSO_3^- formed. This conversion value then serves as the initial concentration for determining the final concentration of HSO_3^- . This structured approach ensures accuracy and efficiency, providing a comprehensive understanding of atmospheric pollutant dissolution and subsequent acid formation in rainwater.

After calculations, we obtain three conversion values for SO_2 , HSO_3^- , and HNO_3 , representing their contributions to H^+ ion formation. The conversions from SO_2 and HSO_3^- collectively contribute to H^+ ions via the SO_2 pathway, while the HNO_3 conversion directly contributes to H^+ ions via the HNO_3 pathway. This distinction highlights the unique contributions of each pollutant to rainwater acidity.

$$\begin{aligned} \text{Total conversion of } H^+ \text{ through } SO_2 &= \text{conversion} \\ &\text{of } SO_2 + \text{conversion of } HSO_3^- \\ &= 0.995 + 0.00003 = 0.99503 \end{aligned}$$

Vinothkumar et al.,

Total conversion of H^+ through $HNO_3 =$ conversion of $HNO_3 = 0.9999$

After calculating the conversions X , the n_i term in our model is replaced with the corresponding conversion values are,

$$pH = -\log_{10} \left[\frac{1.004 \times 10^{-6} \times 1000 \times ((0.9950 \times 4.08 \times 10^{-7}) + (0.9999 \times 4.078 \times 10^{-9}))}{2.679 \times 10^{-7}} \right]$$

$$pH = 2.8134$$

After substituting n_i with the conversion factor X , the calculated pH value is 2.8134, reducing the deviation from the reference value by 41.06%, an improvement from the initial 47.3%. This refinement enhances the model's predictive accuracy and alignment in Fig. 2 and 3. The pH value from reference and from calculation at various fall distance were given in Table. 2. The detail methods, calculations, and validating processes to derive an equation for predicting the pH value of rainwater were given in the supplementary information.

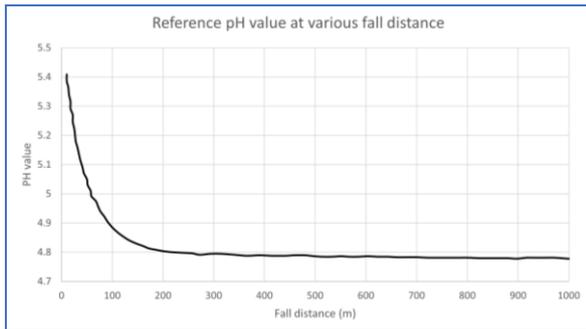


Fig 2: Reference pH value of rain drop at various fall distance [29]

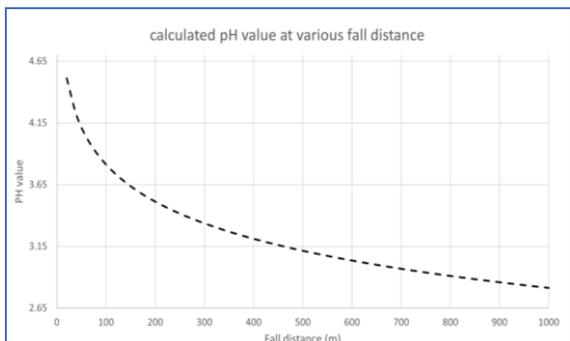


Fig 3: Calculated pH value of rain drops at various fall distance using the derived equation through MATLAB

To enhance accuracy, the final equation incorporates the conversion factor X_i instead of the n_i . The comparative result of before and after refinement of the derived equation has been

represented in the Fig. 4. The results are in a more precise representation of the relationship and further optimizing the equation for predicting rainwater acidity. The refined equation is as follows,

$$pH = -\log_{10} \left[\frac{S_{avg} \cdot h_R \cdot \left(\sum_{j=i} \frac{X_j \cdot C_{i0}}{1 - e^{-\left(\frac{V_{max} \cdot V_{ref}}{D_{iR}} \right)}} \right)}{V_R} \right] \quad 24$$

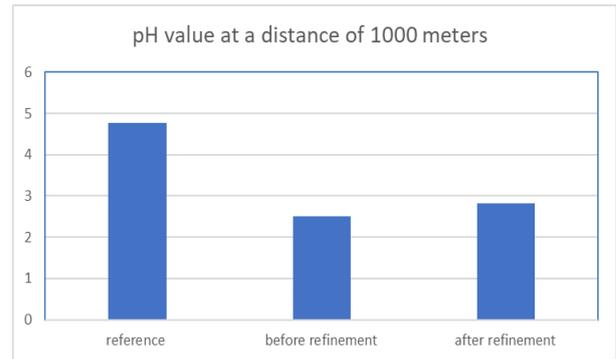


Fig 4: A comparison plot between reference pH, calculated pH before and after modification

Table 1: Diffusion coefficient or diffusivity D_{iR} , of air pollutants in water at atmospheric pressure and given temperature [24]

Air pollutants	10 °C	15 °C	20 °C	25 °C	30 °C	35 °C
Sulphur dioxide	-	-	1.62	1.83	2.07	2.32
Nitrogen dioxide	-	-	1.23	1.40	1.59	-
Carbon dioxide	1.26	1.45	1.67	1.92	2.17	2.47

4. CONCLUSION

In conclusion, our research demonstrates the effectiveness of the derived equation in predicting rainwater pH. Initially, the calculated pH value deviated by 47.3% from the reference value. By substituting the term 'n' with the

conversion factor 'X', we reduced this deviation to 41.06%, which confirms the significant improvement in prediction accuracy. Our method, which considers the rate equations of all pollutants with water and focuses on the forward reaction, provides a foundation for further research. Future studies should account for both forward and backward reactions and variability in fall distance to enhance accuracy. The model's inclusion of various acids ensures its versatility across different geographical locations and environmental contexts. Further research and validation are necessary to evaluate the equation's real-world performance and practical applicability. Overall, our findings contribute to more accurate predictions of rainwater acidity, aiding in the preservation of historical structures and mitigating acid rain's ecological and health impacts. This research advances our understanding and management of acid rain to realize more sustainable future.

Table 2: pH value from reference and from calculation at various fall distances

Sl.No.	Fall distance	pH value from	
		Reference [29]	Calculated
1.	20	5.4083	4.5124
2.	100	5.3150	3.8134
3.	200	5.2033	3.5124
4.	300	5.0916	3.3363
5.	400	4.9916	3.2114
6.	500	4.9033	3.1144
7.	600	4.8300	3.0353
8.	700	4.7999	2.9683
9.	800	4.7950	2.9103
10.	900	4.7883	2.8592
11.	1000	4.7850	2.8134

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