A Nonlinear Dynamical Model to Study the Removal of Gaseous and Particulate Pollutants in a Rain System

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Received: 05.01.2006 **Revised:** 27.02.2007 **Published online:** 05.05.2007

Abstract. An ecological type nonlinear mathematical model is proposed to study the removal of gaseous pollutants and two distinct particulate matters by precipitation scavenging in the atmosphere. The atmosphere during precipitation consists of five interacting phases namely the raindrops phase, the gaseous pollutants phase, the smaller particulate matters phase, the larger particulate matters phase and the absorbed phase of gaseous pollutants. We assume that gaseous pollutants are removed from the atmosphere by the processes of impaction as well as by absorption while particulate matters are assumed to be removed by impaction process. The model is analyzed using stability theory of nonlinear differential equations. It is shown that, under appropriate conditions, the pollutants can be removed from the atmosphere and their removal rates would depend mainly upon the rates of emission of pollutants, rate of rain drops formation and the rate of raindrops falling on the ground. If the rate of precipitation is very high, all the pollutants (gaseous as well as particulate matters) would be removed completely from the atmosphere. A numerical study is also performed to study the dynamics of the model system. The results are found to be in line with the experimental observations published in the literature.

Keywords: nonlinear model, gaseous pollutants, particulate matters, precipitation scavenging, stability.

1 Introduction

The role of monsoon rain in agricultural production, development of watershed, land slides, floods etc. in Indian subcontinent is well known but its very important role in cleaning the polluted environment of industrial cities is less understood. The atmosphere of Kanpur city in Utter Pradesh, India, for example, is highly polluted by various kinds of gaseous pollutants as well as particulate matters of different sizes due to variety of industries, exhausts from vehicular traffic, household waste materials, etc. Gases like SO_2 and NO_2 released from industries, power plants etc. by burning fossil fuels when reach high into the atmosphere, combine with available moisture to form acid rain which is very harmful to the environment. Larger particulate matters ranging from 2.5 microns to 100 microns in diameter, usually comprise smoke and dust from industrial processes, agriculture, construction and road traffic, etc. The smaller particulate matters, less than 2.5 micron in diameter, come from combustion of fossil fuels. Particulate matters may cause acute changes in lung function, respiratory and heart diseases etc. When the level of particulate matters in the air increases up to 200 micrograms per meter cube, daily mortality rate could increase up to 20 percent. Thus, the removal of pollutants from the atmosphere is the key question. Precipitation scavenging provides an important mechanism for the removal of these pollutants from the atmosphere, in which atmospheric gases/ particulate matters are absorbed/trapped in raindrops falling on the ground. The phenomenon of absorption/impaction of these pollutants by raindrops is the key step in removal of gaseous pollutants and particulate matters. The removal of particulate matters may, however, be dependent on their size and shape.

In experimental studies, it has been shown that the pollutants are removed from the atmosphere by precipitation process [1–9]. In particular, Sharma et al. [9] measured the concentration of suspended particulate matters in Kanpur city, UP, India and found considerable decrease in their concentrations during monsoon. Davies [3] studied the removal of SO_2 by precipitation in an industrial area of Sheffield, UK and found significant reduction in its concentration after rain.

Some mathematical investigations have also been made to study the dynamics of removal of pollutants in a rain system [2,10–19]. Hales [11] presented some fundamentals for the general analysis of precipitation scavenging emphasizing the importance of reversible phenomena. Chang [2] has derived wet removal coefficient for nitric acid vapour in rain and snow systems and parameterized them in terms of precipitation rate under a number of approximations. Since the concentration of pollutants in gaseous phase directly influences the concentration of pollutants in raindrops phase, it is, therefore, necessary to consider simultaneously the coupled process of gas phase depletion and aqueous phase accumulation of pollutants. Kumar [12] has given an Eulerian model to describe the simultaneous process of trace gas removal from the atmosphere and absorption of these gases in raindrops by considering the precipitation scavenging of these gases present below the clouds. Pandis and Seinfeld [17] have studied the interactions between equilibration process and wet or dry deposition. They considered three cases to obtain useful insight into the interaction and deposition process. The first case was related to a gas phase species which can be reversibly transferred to aerosol phase. In the second phase, the gas phase species in presence of droplets of liquid water (fog) was transferred reversibly to the aqueous phase. In the third case, two gases reacted to give a volatile aerosol phase. They have found interesting relationship in all the three cases and, in particular, found in the second phase (i.e. in the fog episode), the deposition of gaseous species increased by as much as three times. The process of trace gases scavenging is reversible in nature and the phenomenon of absorption and desorption may cause a redistribution of pollutants in the atmosphere [20–22]. Slinn [22] has analytically considered the redistribution of gas plume caused by reversible washout and presented solution in some simple cases. Fisher [20] has studied the transport and removal of sulfur dioxide in rain using a simple model which combines the microphysics of absorption and chemical transformation of sulfur dioxide in cloud with the dynamics of the air motion and obtained appropriate one dimensional solution of the governing equations. Naresh [15] studied the effect of precipitation scavenging on the unsteady state dispersion of a reactive gaseous air pollutant emitted from a time dependent point source, assuming uniform distribution of raindrops in the atmosphere which absorb the pollutant and remove it by their fall on the ground.

It may be noted that the dispersion of air pollutants and their removal from the atmosphere by precipitation has been modelled mainly using coupled linear convective diffusion equations for the gaseous and particulate phase and the absorbed pollutant in the raindrops phase by taking into account uniform distribution of raindrops. However, it may be pointed out that in real situations, during rain, the number density of the raindrops changes as the intensity of precipitation increases. This change in number density affects the interaction process between raindrops and gaseous pollutants as well as with particulate matters (aerosols), making the phenomenon nonlinear and should, therefore, be taken into account in the model. The main mechanism for removal of gaseous pollutants is through the falling of raindrops and the removal term, in general, due to precipitation, is proportional to the concentration of the absorbed pollutant as well as to the number density of raindrops in the atmosphere. A little attention has been paid to study the problem of removal of pollutants by precipitation using nonlinear models though it involves nonlinear interactions of various phases in the atmosphere [23–25]. For example, Naresh et al. [24] presented a nonlinear mathematical model to study the removal of primary and secondary pollutants from the atmosphere of an industrial city by rain.

In view of the above considerations, in this paper we propose a nonlinear mathematical model to study the removal of gaseous pollutants and two distinct particulate matters of different size from the stable atmosphere of a polluted region by precipitation scavenging. Our objective is to analyze the proposed nonlinear model to see the effect of precipitation scavenging on the equilibrium level of pollutants in the atmosphere by using stability theory [23, 24, 26, 27]. A numerical study of the model is also performed to see the role of key parameters on the removal process. The model can further be generalized by including diffusion and convection terms if atmospheric conditions such as wind, temperature inversion, topography of the terrain etc. are also to be taken into account.

2 Mathematical model

Consider the stable atmosphere of a polluted region where rain is taking place. We assume that there exist five interacting phases: the raindrops, the gaseous pollutants, the smaller particulate matters, the larger particulate matters and the gaseous pollutants absorbed in raindrops in the atmosphere.

It is pointed out here that during high intensity of rain, the contact time of gaseous pollutants phase with raindrops phase is very short before falling on the ground and therefore it may be reasonable to assume that the interaction between these phases is governed by simple law of mass action i.e. bilinear interaction. A similar situation also arises in the case of interaction of particulate matters with raindrops phase. The gaseous pollutants are removed by precipitation as well as by natural factors such as effect of gravity, interactions with plant leaves, buildings, walls, roofs, etc. while the removal of particulate matters takes place by the processes of impaction and entrapment by raindrops and by natural factors as well. The removal rates of gaseous pollutants and particulate matters are assumed to be proportional to their respective cumulative concentrations. Further, it is also assumed that a fraction of gaseous pollutants absorbed in the raindrops may re-enter into the atmosphere by the reversible process. Here in the modeling process, our aim is to emphasize the removal of pollutants by precipitation rather than their chemical kinetics involved in the interaction processes.

Let $C_r(t)$ be the number density of raindrops in the atmosphere, $C(t), C_{p1}(t)$ and $C_{p2}(t)$ be the cumulative concentrations of gaseous pollutants, smaller particulate matters and larger particulate matters respectively. It is assumed that the raindrops may get depleted naturally and also by interaction with the gaseous pollutants as this is proportional to the raindrops density as well as the concentration of gaseous pollutants, $q(t)$ is the rate of formation of raindrops assumed to be constant (say q), r_0 is the natural depletion rate coefficient of raindrops, r is the depletion rate coefficient of raindrops due to interaction with C.

Thus, the dynamics of raindrops density is assumed to be governed by the following equation,

$$\frac{dC_r}{dt} = q(t) - r_0 C_r - r C_r C. \tag{1}$$

To write the other equations, it is assumed that $Q(t), Q_1(t)$ and $Q_2(t)$ are the emission rates of gaseous pollutants, smaller particulate matters and larger particulate matters respectively with their natural depletion rates $\delta C, \delta_1 C_{p1}$ and $\delta_2 C_{p2}$. It is also assumed that the growth rate of larger particulate matters is further enhanced by the agglomeration of smaller particulates with a rate β [28]. Further, the absorption/impaction of these pollutants is proportional to the number density of raindrops as well as the cumulative concentrations of respective pollutants (i.e. $\alpha C C_r, \alpha_1 C_{p1} C_r$ and $\alpha_2 C_{p2} C_r$). The gaseous pollutants in the absorbed phase may be removed by the rate kC_a and a fraction of it (i.e. $\theta k C_a$) may re-enter into the atmosphere by recycling process. It is also assumed that the removal of gaseous pollutants in the absorbed phase is proportional to its concentration in absorbed phase and the number density of raindrops (i.e. $\nu C_r C_a$) and a fraction of it (i.e. $\pi \nu C_r C_a$) may also re-enter into the atmosphere by reversible process to increase the concentration of gaseous pollutants in the atmosphere. The constants $0 \le \theta,\ \pi \le 1$ are the reversible rate coefficients.

In view of the above, the dynamics of these phases can be written by the following system of differential equations,

$$\frac{dC}{dt} = Q(t) - \delta C - \alpha C C_r + \theta k C_a + \pi \nu C_r C_a, \tag{2}$$

$$\frac{dC_{p1}}{dt} = Q_1(t) - (\delta_1 + \beta) C_{p1} - \alpha_1 C_{p1} C_r, \tag{3}$$

$$\frac{dC_{p2}}{dt} = Q_2(t) + \beta C_{p1} - \delta_2 C_{p2} - \alpha_2 C_{p2} C_r, \tag{4}$$
$$\frac{dC_a}{dt} = \alpha C C_r - k C_a - \nu C_r C_a, \tag{5}$$
$$C_r(0) \geq 0, \quad C(0) \geq 0, \quad C_{p1}(0) \geq 0, \quad C_{p2}(0) \geq 0, \quad C_a(0) \geq 0.$$

In equations (2)–(5), the constants $\delta, \delta_1, \delta_2$ and k are natural removal rate coefficients of C, C_{p1}, C_{p2} and C_a respectively, α is the absorption rate coefficient of C due to interaction with C_r, α_1 and α_2 are the impaction rate coefficients of C_{p1} and C_{p2} respectively and ν is the removal rate coefficient of absorbed phase. It may be noted that when the distribution of C_r is constant then the model reduces to the linearised model similar to that given by Slinn [22] and Kumar [12] without diffusion and convection.

In the following, we analyze the model (1)–(5) using the stability theory of differential equations. We need bounds of dependent variables involved in the model [29–32]. For this, we state the region of attraction as follows.

The set

$$\Omega = \Big\{ (C_r, C, C_{p1}, C_{p2}, C_a) \colon \ 0 \leq C_r \leq \frac{q}{r_0}, \ 0 \leq C + C_a \leq \frac{Q}{\delta_m}, \\ 0 \leq C_{p1} \leq \frac{Q_1}{\delta_1 + \beta}, \ 0 \leq C_{p2} \leq \frac{Q_n}{\delta_2} \Big\}$$

attracts all solutions initiating in the interior of the positive octant, where

$$\delta_m = \min\big\{\delta, (1-\theta)k\big\} \quad \text{and} \quad Q_n = Q_2 + \frac{\beta Q_1}{\delta_1 + \beta}.$$

3 Stability analysis

Now we analyze the model (1)–(5) under the following two cases. The first case represents the emission of pollutants in the atmosphere with constant rate, for example, by stacks emitting continuously, whereas the second case corresponds to the situation when the pollutants are emitted in the atmosphere by an instantaneous source.

1. $Q(t) = Q, \ Q_1(t) = Q_1, \ Q_2(t) = Q_2$ and $q(t) = q$ (constant emission).
2. $Q(t) = 0, \ Q_1(t) = 0, \ Q_2(t) = 0$ and $q(t) = q$ (instantaneous emission).

3.1 Case I: constant emission $Q(t) = Q, \ Q_1(t) = Q_1, \ Q_2(t) = Q_2$ and $q(t) = q$

In this case, the model has only one non-negative equilibrium namely $E^*(C_r^*, C^*, C_{p1}^*, C_{p2}^*, C_a^*)$ where $C_r^*, C^*, C_{p1}^*, C_{p2}^*$, and C_a^* are the positive solutions of the following system of algebraic equations,

$$C_r = \frac{q}{r_0 + rC}, \tag{6}$$
$$C = \frac{Q(k + \nu C_r)}{\delta k + \big(\delta\nu + (1-\theta)\alpha k\big)C_r + (1-\pi)\alpha\nu C_r^2} = f(C_r), \tag{7}$$

$$C_{p1} = \frac{Q_1}{\delta_1 + \beta + \alpha_1 C_r}, \tag{8}$$

$$C_{p2} = \frac{Q_2 + \beta C_{p1}}{\delta_2 + \alpha_2 C_r}, \tag{9}$$

$$C_a = \frac{\alpha C C_r}{k + \nu C_r}. \tag{10}$$

From equation (6), let $F(C_r) = 0$ where

$$F(C_r) = q - r_0 C_r - r C_r C \tag{11}$$

which implies $F(0) = q > 0$ and $F(\frac{q}{r_0}) < 0$. Also, $F'(C_r) = -[r_0 + r\{C_r f'(C_r) + f(C_r)\}] < 0$, provided the following condition holds.

$$r_0 + r\big\{C_r f'(C_r) + f(C_r)\big\} > 0. \tag{12}$$

Thus, $F(C_r) = 0$ has exactly one root (say C_r^*) between 0 and $\frac{q}{r_0}$ under condition (12). Using C_r^*, the values of C^*, C_{p1}^*, C_{p2}^* and C_a^* can be found from equations (7), (8), (9) and (10) respectively.

It may also be noted from equations (7)–(10), that $C, C_{p1}, C_{p2}, C_a \to 0$ as $C_r \to \infty$ showing that all the pollutants would be removed completely from the atmosphere, if the number density of raindrops is very high.

Now we check the characteristics of various phases with respect to parameter q, the rate of formation of raindrops.

(i) **Variation of C_r with q.** Differentiating equation (6) with respect to q and using assumption (12), we get $\frac{dC_r}{dq} > 0$. Thus C_r increases with increase in q.

(ii) **Variation of C with q.** From equation (7), we get $\frac{dC}{dC_r} < 0$ and since $\frac{dC_r}{dq} > 0$, it follows that $\frac{dC}{dq} < 0$. Therefore, C decreases with increase in q.

(iii) **Variation of C_{p1} with q.** From equation (8), we have $\frac{dC_{p1}}{dC_r} < 0$ and since $\frac{dC_r}{dq} > 0$, therefore $\frac{dC_{p1}}{dq} < 0$. Thus C_{p1} decreases with increase in q.

(iv) **Variation of C_{p2} with q.** From equation (9), it can be easily seen that $\frac{dC_{p2}}{dq} < 0$ showing that C_{p2} decreases with increase in q.

Hence, as the rate of raindrops formation q increases i.e. as the precipitation intensity increases, the cumulative concentrations of pollutants (gaseous as well as particulate matters) decrease and these may be removed completely for very large q under certain conditions. It is also noted that, if the coefficients $\alpha, \alpha_1, \alpha_2$ are so large that $\frac{dC}{dt} < 0$, $\frac{dC_{p1}}{dt} < 0$, $\frac{dC_{p2}}{dt} < 0$, all the pollutants will be removed from the atmosphere. Also for large ν, $\frac{dC_a}{dt} < 0$ and the formation of absorbed phase is very transient and it may not exist.

To study the stability behavior of the equilibrium, we propose the following theorems.

Theorem 1. *Let the following inequalities hold,*

$$\left[rC_r^* + (\alpha C^* - \pi\nu C_a^*)\right]^2 < \frac{q}{3C_r^*}(\delta + \alpha C_r^*), \tag{13}$$

$$\frac{27}{4}\,\frac{(\theta k+\pi\nu C_r^*)^2}{\delta+\alpha C_r^*} < (k+\nu C_r^*)^2 \min\left\{\frac{q}{C_r^*(\alpha C^*-\nu C_a^*)^2}, \frac{4(\delta+\alpha C_r^*)}{3(\theta k+\pi\nu C_r^*)^2}\right\}, \tag{14}$$

then E^* is locally asymptotically stable.

See Appendix A for proof.

Theorem 2. *Let the following inequalities hold,*

$$\left[r\frac{q}{r_0} + (\alpha C^* - \pi\nu C_a^*)\right]^2 < \frac{q}{3C_r^*}\delta, \tag{15}$$

$$\frac{27}{4}\,\frac{(\theta k+\pi\nu \frac{q}{r_0})^2}{\delta} < (k+\nu C_r^*)^2 \min\left\{\frac{q}{C_r^*(\alpha+\nu)^2\left(\frac{Q}{\delta_m}\right)^2}, \frac{4\delta}{3\left(\theta k+\pi\nu \frac{q}{r_0}\right)^2}\right\}, \tag{16}$$

then E^* is nonlinearly asymptotically stable.

See Appendix B for proof.

These theorems imply that the concentration of the pollutants, gaseous as well as particulate matters, in the atmosphere decreases with increase in the rate of precipitation and removal rate coefficients under certain conditions.

Remark 1. *From equations* (13) *and* (15)*, we note that if the depletion of raindrops (r) due to gaseous pollutants is very small and the corresponding removal δ due to natural factors is very high, the possibility of satisfying the conditions increases.*

Remark 2. *If θ and π both are equal to zero, then the conditions* (14) *and* (16) *are satisfied automatically.*

3.2 Case II: instantaneous emission, $Q(t) = 0,\ Q_1(t) = 0,\ Q_2(t) = 0$ and $q(t) = q$

In this case, the model has only one non-negative equilibrium $E_0(\frac{q}{r_0}, 0, 0, 0, 0)$ in $C_r - C - C_{p1} - C_{p2} - C_a$ space. The existence of E_0 is obvious.

By computing the variational matrix corresponding to E_0, it can be easily shown that E_0 is locally asymptotically stable.

We propose the following theorem to check the global stability character of E_0.

Theorem 3. *If $C_r(0) > 0$, then E_0 is globally asymptotically stable with respect to the non-negative octant.*

Proof. From equation (1), $\frac{dC_r}{dt} \le q - r_0 C_r$. From this we get $\lim\limits_{t\to\inf} \sup C_r(t) \le \frac{q}{r_0}$. Again from equations (2) and (5), we have,

$$\begin{aligned}\frac{dC}{dt} + \frac{dC_a}{dt} &= -\delta C - (1-\theta)kC_a - (1-\pi)\nu C_r C_a \\ &\le -\delta C - (1-\theta)kC_a \le -\delta_m(C + C_a),\end{aligned}$$

where $\delta_m = \min\{\delta, (1-\theta)k\}$.

We find that $C(t) + C_a(t) \le \{C(0) + C_a(0)\} \exp(-\delta_m t)$ and hence the system is dissipative. Using comparison theorem, it can be shown that,

$$\lim_{t\to\inf} \sup C(t) = \lim_{t\to\inf} \sup C_a(t) = 0.$$

Similarly from equations (3) and (4) we have,

$$\lim_{t\to\inf} \sup C_{p1}(t) = 0 \quad \text{and} \quad \lim_{t\to\inf} \sup C_{p2}(t) = 0.$$

Thus, in the limit $C_r(t)$ tends to q/r_0 and since $C_r(0) > 0$, the theorem follows. □

This theorem implies that in the case of instantaneous emission, the gaseous pollutants and both the particulate matters are washed out completely from the atmosphere by rain with the number density of raindrops remaining at its equilibrium. The time taken for removal will depend upon the rate of raindrops formation and removal rate coefficients.

4 Numerical simulation

In this section we present the results of numerical analysis of the model (1)–(5). Consider the following set of parameters,

$$\begin{array}{llllll} Q = 5.0, & Q_1 = 3.0, & Q_2 = 2.0, & q = 10, & r = 0.0003, & r_0 = 0.2, \\ \delta = 0.25, & \delta_1 = 0.30, & \delta_2 = 0.35, & k = 0.40, & \theta = 0.00003, & \pi = 0.00002, \\ \alpha = 0.70, & \alpha_1 = 0.55, & \alpha_2 = 0.60, & \beta = 0.05, & \nu = 0.55. & \end{array}$$

The equilibrium E^* is given by

$$\begin{array}{lll} C_r^* = 49.989361, & C^* = 0141876, & C_{p1}^* = 0.107742, \\ C_{p2}^* = 0.066089, & C_a^* = 0.177981. & \end{array}$$

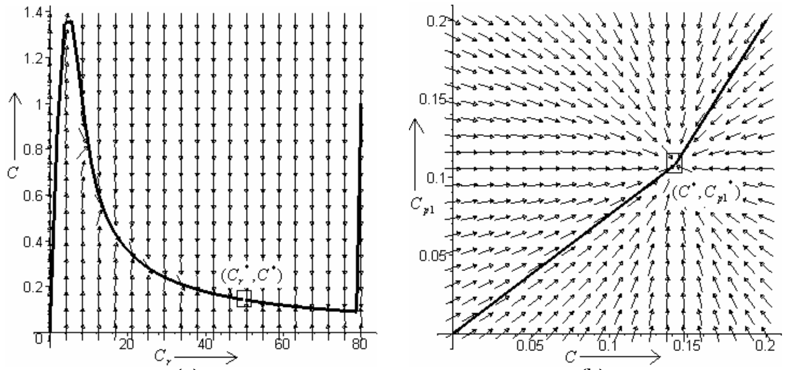
Eigen values corresponding to E^* are obtained as

$$-30.343616, \ -27.844148, \ -35.245270, \ -27.891474, \ -0.200000.$$

Since all the eigen values corresponding to E^* are negative, therefore E^* is locally asymptotically stable.

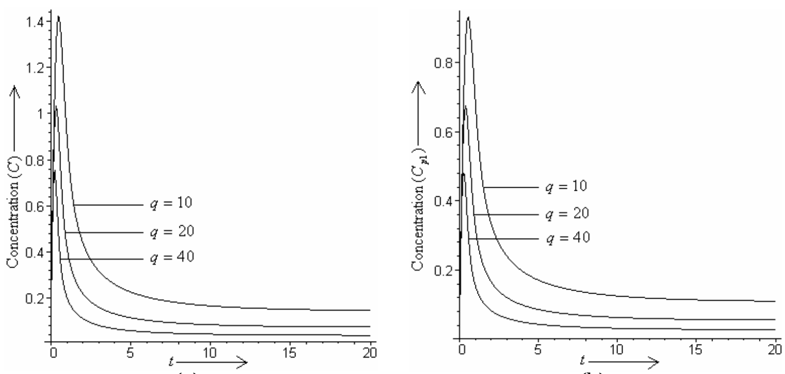
The nonlinear stability behavior of E^* in $C_r - C$ and $C - C_{p1}$ plane is shown in the Fig. 1(a) and Fig. 1(b) respectively. It can also be checked that for the above set of parameters, the local as well as nonlinear stability conditions are satisfied. In Figs. 2, 3, the variation of cumulative concentration of gaseous pollutants (C), particulate matters (C_{p1} and C_{p2}) and absorbed phase (C_a) of gaseous pollutants respectively with time t is shown for different values of raindrops formation i.e. $q = 10, 20, 40$. From these figures, it is seen that the cumulative concentrations of gaseous pollutants (C), particulate matters (C_{p1} and C_{p2}) and that of gaseous pollutants in absorbed phase (C_a) decrease as

q increases. Fig. 4 shows the variation of cumulative concentration of particulate matters C_{p1}, C_{p2} and absorbed phase (C_a) of gaseous pollutants with time t for different values of $\alpha_1,\ \alpha_2$ and ν respectively. From these figures, it can be seen that the cumulative concentrations of particulate matters C_{p1}, C_{p2} and concentration of gaseous pollutants in absorbed phase (C_a) decrease with increases in respective removal parameters. Thus we note that with the increase in the removal rate coefficients the pollutants are significantly removed from the atmosphere by rain. These results are qualitatively similar to the experimental observations as has already been pointed out in the Introduction. In the Table 1, the variation of equilibrium values of E^* is shown for different values of raindrops formation q. From this, it is clear that the densities of rain drops increase while the cumulative concentrations of gaseous pollutants (C), particulate matters C_{p1}, C_{p2} and concentration of gaseous pollutants in absorbed phase (C_a) decrease with increase in rain drops formation q. In the Tables 2, 3 and 4, it is shown that the concentration of particulate matters C_{p1}, C_{p2} and gaseous pollutants in absorbed phase (C_a) decreases with increase in removal parameters.



(a) (b)

Fig. 1. Nonlinear asymptotic stability in $C_r - C$ plane (a); in $C - C_{p1}$ plane (b).



(a) (b)

Fig. 2. Variation of C (a); variation of C_{p1} (b) with time t for different values of q.

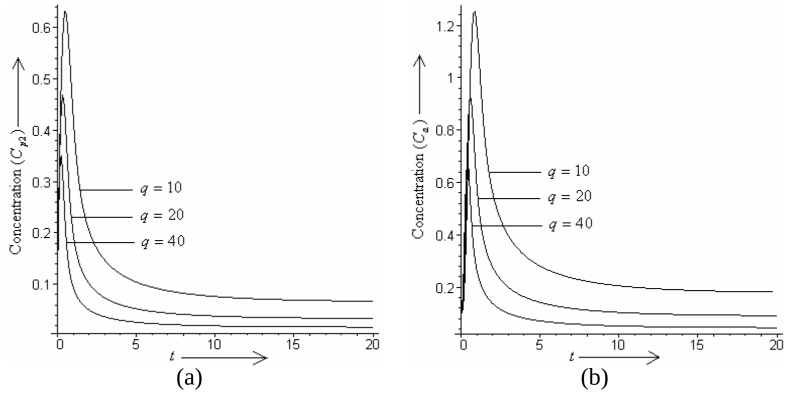


Fig. 3. Variation of C_{p2} (a); variation of C_a (b) with time t for different values of q.

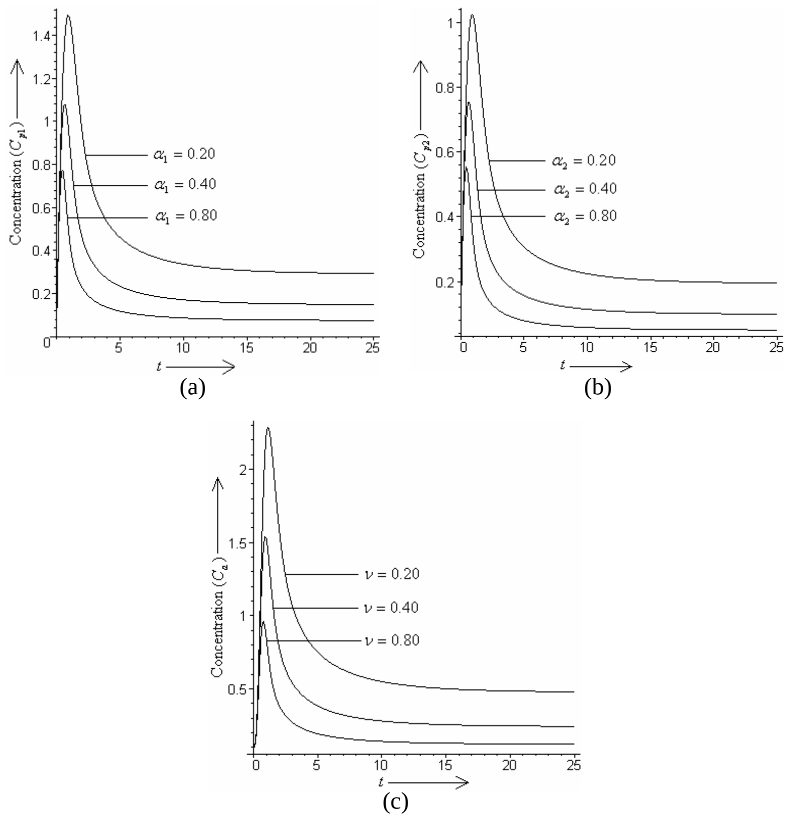


Fig. 4. Variation of C_{p1} with time t for different values of α_1 (a); C_{p2} with time t for different values of α_2 (b); C_a with time t for different values of ν (c).

Table 1. Variation of equilibrium values with q

q	C_r^*	C^*	C_{p1}^*	C_{p2}^*	C_a^*
10	49.989361	0.141876	0.107742	0.066089	0.177981
15	74.989336	0.094802	0.072125	0.044187	0.119498
20	99.989323	0.071183	0.054206	0.033188	0.089942
25	124.989316	0.056986	0.043419	0.026573	0.072108
30	149.989311	0.047510	0.036212	0.022157	0.060175

Table 2. Variation of C_{p1}^* with α_1

α_1	0.55	0.60	0.65	0.70	0.75
C_{p1}^*	0.107742	0.098867	0.091343	0.084883	0.079276

Table 3. Variation of C_{p2}^* with α_2

α_2	0.60	0.65	0.70	0.75	0.80
C_{p2}^*	0.066089	0.061059	0.056741	0.052993	0.049710

Table 4. Variation of C_a^* with ν

ν	0.55	0.60	0.65	0.70	0.75
C_a^*	0.177981	0.163344	0.150932	0.140273	0.131020

5 Conclusion

In this paper, we have proposed and analyzed a nonlinear dynamical model to study the removal of gaseous pollutants and two distinct particulate matters of different size from the atmosphere of a city by rain. It has been assumed that the removal of gaseous pollutants takes place by the process of absorption by raindrops falling on the ground while the removal of particulate matters by the processes of impaction and entrapment by falling raindrops. It has been shown qualitatively and numerically that when the pollutants are emitted in the atmosphere by an instantaneous source, all the pollutants from the atmosphere would be completely removed by precipitation scavenging. When the pollutants are emitted at a constant rate, these pollutants can still be washed out from the atmosphere under appropriate conditions and the rate of removal would depend upon the rate of emission of pollutants, the rate of raindrops formation and removal parameters. The equilibrium level of gaseous pollutants and that of particulate matters in the atmosphere is much smaller after rain than its corresponding value before rain. It is also noted that for large precipitation rate, the equilibrium concentration of pollutants reduces considerably in the atmosphere. It is clear that the rate of removal of larger particulate matters is greater than that of smaller particulate matters as expected. The results are qualitatively similar to the results predicted by linear models and are qualitatively in line with experimental observations.

From the analysis, it may be speculated that raindrops, fog droplets or even externally introduced species (liquid) can be very effective in neutralizing the effect of gaseous pollutants in the atmosphere. The analysis also suggests a mechanism by which toxic gases leaked in the atmosphere due to accidental discharges from storage tanks etc. can be removed by introducing artificially the liquid phase in the atmosphere which can interact with toxic material to neutralize its effect.

Acknowledgement

Authors are thankful to the reviewers for their constructive suggestions which helped us in finalizing the paper.

Appendix A: Proof of the Theorem 1

To establish the local stability of E^*, let us consider the following positive definite function,

$$V = \frac{1}{2}(C_{r1}^2 + k_1 C_1^2 + k_2 C_{p11}^2 + k_3 C_{p21}^2 + k_4 C_{a1}^2), \tag{A.1}$$

where $C_{r1},\ C_1,\ C_{p11},\ C_{p21}$ and C_{a1} are the small perturbations about E^* as

$$\begin{aligned} C_r &= C_r^* + C_{r1}, \quad C = C^* + C_1, \quad C_{p1} = C_{p1}^* + C_{p11}, \\ C_{p2} &= C_{p2}^* + C_{p21}, \quad C_a = C_a^* + C_{a1}. \end{aligned}$$

Differentiating (A.1) with respect to t we get

$$\dot{V} = C_{r1}\dot{C}_{r1} + k_1 C_1 \dot{C}_1 + k_2 C_{p11}\dot{C}_{p11} + k_3 C_{p21}\dot{C}_{p21} + k_4 C_{a1}\dot{C}_{a1}. \tag{A.2}$$

The linearized system of model equations (1)–(5) corresponding to E^* is

$$\begin{aligned} \dot{C}_{r1} &= -\frac{q}{C_r^*}C_{r1} - rC_r^* C_1, \\ \dot{C}_1 &= -(\alpha C^* - \pi\nu C_a^*)C_{r1} - (\delta + \alpha C_r^*)C_1 + (\theta k + \pi\nu C_r^*)C_{a1}, \\ \dot{C}_{p11} &= -\alpha_1 C_{p1}^* C_{r1} - (\delta_1 + \beta + \alpha_1 C_r^*)C_{p11}, \\ \dot{C}_{p21} &= -\alpha_2 C_{p2}^* C_{r1} + \beta C_{p11} - (\delta_2 + \alpha_2 C_r^*)C_{p21}, \\ \dot{C}_{a1} &= (\alpha C^* - \nu C_a^*)C_{r1} + \alpha C_r^* C_1 - (k + \nu C_r^*)C_{a1}. \end{aligned}$$

Now from (A.2), we have

$$\begin{aligned}\dot{V} = &-\frac{q}{C_r^*}C_{r1}^2 - k_1(\delta+\alpha C_r^*)C_1^2 - k_2(\delta_1+\beta+\alpha_1 C_r^*)C_{p11}^2 - k_3(\delta_2+\alpha_2 C_r^*)C_{p21}^2 \\ &- k_4(k+\nu C_r^*)C_{a1}^2 - \left[rC_r^* + k_1(\alpha C^* - \pi\nu C_a^*)\right]C_{r1}C_1 - k_2\alpha_1 C_{p1}^* C_{r1}C_{p11} \\ &- k_3\alpha_2 C_{p2}^* C_{r1}C_{p21} + k_4(\alpha C^* - \nu C_a^*)C_{r1}C_{a1} \\ &+ \left[k_1(\theta k + \pi\nu C_r^*) + k_4\alpha C_r^*\right]C_1 C_{a1} + \beta k_3 C_{p11}C_{p21}.\end{aligned}$$

$\dot{V}$ will be negative definite provided the following conditions hold,

$$\left[rC_r^* + k_1(\alpha C^* - \pi\nu C_a^*)\right]^2 < \frac{q}{3C_r^*}k_1(\delta_0 + \delta + \alpha C_r^*), \tag{A.3}$$

$$k_2[\alpha_1 C_{p1}^*]^2 < \frac{q}{2C_r^*}(\delta_1 + \beta + \alpha_1 C_r^*), \tag{A.4}$$

$$k_3[\alpha_2 C_{p2}^*]^2 < \frac{q}{2C_r^*}(\delta_2 + \alpha_2 C_r^*), \tag{A.5}$$

$$k_4\left[(\alpha C^* - \nu C_a^*)\right]^2 < \frac{q}{3C_r^*}(k + \nu C_r^*), \tag{A.6}$$

$$k_1(\theta k + \pi\nu C_r^*)^2 < \frac{4}{9}k_4(\delta + \alpha C_r^*)(k + \nu C_r^*), \tag{A.7}$$

$$k_4(\alpha C_r^*)^2 < \frac{4}{9}k_1(\delta + \alpha C_r^*)(k + \nu C_r^*), \tag{A.8}$$

$$k_3\beta^2 < k_2(\delta_1 + \beta + \alpha_1 C_r^*)(\delta_2 + \alpha_2 C_r^*). \tag{A.9}$$

Now choosing

$$\begin{aligned}&k_1 = 1, \quad 0 < k_2 < \frac{q(\delta_1 + \beta + \alpha_1 C_r^*)}{2C_r^*(\alpha_1 C_{p1}^*)^2}, \\ &0 < k_3 < \frac{q(\delta_2 + \alpha_2 C_r^*)}{2C_r^*}\min\left\{\frac{(\delta_1 + \beta + \alpha_1 C_r^*)^2}{(\alpha_1 C_{p1}^*)^2}, \frac{1}{(\alpha_2 C_{p2}^*)^2}\right\}\end{aligned}$$

the above equations reduce to,

$$\left[rC_r^* + (\alpha C^* - \pi\nu C_a^*)\right]^2 < \frac{q}{3C_r^*}(\delta + \alpha C_r^*) \tag{A.10}$$

$$\frac{27}{4}\frac{(\theta k+\pi\nu C_r^*)^2}{(\delta+\alpha C_r^*)} < (k+\nu C_r^*)^2 \min\left\{\frac{q}{C_r^*(\alpha C^*-\nu C_a^*)^2}, \frac{4(\delta+\alpha C_r^*)}{3(\theta k+\pi\nu C_r^*)^2}\right\}. \tag{A.11}$$

Thus under the above conditions $\dot{V}$ will be negative definite showing that V is a Lyapunov function and hence the theorem.

Appendix B: Proof of the Theorem 2

Consider the following positive definite function about E^*,

$$U = \frac{1}{2}\big[(C_r - C_r^*)^2 + m_1(C - C^*)^2 + m_2(C_{p1} - C_{p1}^*)^2 + m_3(C_{p2} - C_{p2}^*)^2 + m_4(C_a - C_a^*)^2\big]. \tag{B.1}$$

Differentiating (B.1) with respect to t we get,

$$\dot{U} = (C_r - C_r^*)\dot{C}_r + m_1(C - C^*)\dot{C} + m_2(C_{p1} - C_{p1}^*)\dot{C}_{p1} + m_3(C_{p2} - C_{p2}^*)\dot{C}_{p2} + m_4(C_a - C_a^*)\dot{C}_a,$$

$$\begin{aligned}\dot{U} = {} & (C_r - C_r^*)(q - r_0C_r - rC_rC) \\ & + m_1(C - C^*)(Q - \delta C - \alpha CC_r + \theta kC_a + \pi\nu C_rC_a) \\ & + m_2(C_{p1} - C_{p1}^*)\big(Q_1 - (\delta_1 + \beta)C_{p1} - \alpha_1 C_{p1}C_r\big) \\ & + m_3(C_{p2} - C_{p2}^*)(Q_2 + \beta C_{p1} - \delta_2 C_{p2} - \alpha_2 C_{p2}C_r) \\ & + m_4(C_a - C_a^*)(\alpha CC_r - kC_a - \nu C_rC_a).\end{aligned}$$

After some algebraic manipulations, it can be written as

$$\begin{aligned}\dot{U} = {} & -m_1\alpha C_r(C - C^*)^2 - \frac{q}{C_r^*}(C_r - C_r^*)^2 - m_1\delta(C - C^*)^2 \\ & - m_2(\delta_1 + \beta + \alpha_1 C_r^*)(C_{p1} - C_{p1}^*)^2 - m_3(\delta_2 + \alpha_2 C_r^*)(C_{p2} - C_{p2}^*)^2 \\ & - m_4(k + \nu C_r^*)(C_a - C_a^*)^2 - \big[rC_r + m_1(\alpha C^* - \pi\nu C_a^*)\big](C_r - C_r^*)(C - C^*) \\ & - m_2\alpha_1 C_{p1}(C_r - C_r^*)(C_{p1}^* - C_{p1}^*) - m_3\alpha_2 C_{p2}(C_r - C_r^*)(C_{p2} - C_{p2}^*) \\ & + \big[m_1(\theta k + \pi\nu C_r) + m_4\alpha C_r^*\big](C - C^*)(C_a - C_a^*) \\ & + \beta m_3(C_{p1} - C_{p1}^*)(C_{p2} - C_{p2}^*) + m_4(\alpha C - \nu C_a)(C_r - C_r^*)(C_a - C_a^*).\end{aligned}$$

$\dot{U}$ will be negative definite provided the following conditions hold,

$$\big[rC_r + m_1(\alpha C^* - \pi\nu C_a^*)\big]^2 < \frac{q}{3C_r^*}m_1\delta, \tag{B.2}$$

$$m_2[\alpha_1 C_{p1}]^2 < \frac{q}{2C_r^*}(\delta_1 + \beta + \alpha_1 C_r^*), \tag{B.3}$$

$$m_3[\alpha_2 C_{p2}]^2 < \frac{q}{2C_r^*}(\delta_2 + \alpha_2 C_r^*), \tag{B.4}$$

$$m_4\big[(\alpha C - \nu C_a)\big]^2 < \frac{q}{2C_r^*}(k + \nu C_r^*), \tag{B.5}$$

$$m_1(\theta k + \pi\nu C_r)^2 < \frac{4}{9}m_4\delta(k + \nu C_r^*), \tag{B.6}$$

$$m_4(\alpha C_r^*)^2 < \frac{4}{9}m_1\delta(k + \nu C_r^*), \tag{B.7}$$

$$m_3\beta^2 < m_2(\delta_1 + \beta + \alpha_1 C_r^*)(\delta_2 + \alpha_2 C_r^*). \tag{B.8}$$

Now choosing

$$m_1 = 1, \quad 0 < m_2 < \frac{q(\delta_1 + \beta + \alpha_1 C_r^*)}{2C_r^* \left(\frac{\alpha_1 Q_1}{\delta_1+\beta}\right)^2},$$
$$0 < m_3 < \frac{q(\delta_2 + \alpha_2 C_r^*)}{2C_r^*} \min\left\{ \frac{(\delta_1 + \beta + \alpha_1 C_r^*)^2}{\left(\frac{\alpha_1 Q_1}{\delta_1+\beta}\right)^2}, \frac{1}{\left(\frac{\alpha_2 Q_n}{\delta_2}\right)^2} \right\}$$

the above equations reduce to,

$$\left[r\frac{q}{r_0} + (\alpha C^* - \pi\nu C_a^*)\right]^2 < \frac{q}{3C_r^*}\delta, \tag{B.9}$$

$$\frac{27}{4}\frac{(\theta k + \pi\nu\frac{q}{r_0})^2}{\delta} < (k + \nu C_r^*)^2 \min\left\{ \frac{q}{C_r^*(\alpha+\nu)^2\left(\frac{Q}{\delta_m}\right)^2}, \frac{4\delta}{3\left(\theta k + \pi\nu\frac{q}{r_0}\right)^2} \right\}. \tag{B.10}$$

Under conditions (B.9) and (B.10), $\dot{U}$ will be negative definite showing that U is a Liapunov's function and hence the theorem.

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