On the Transfer of Dense Gases from a Forest Sub-Canopy into the Wind Field Above.

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Prepared for submission to Atmospheric Environment

Abstract

The need to anticipate high risk of exposure following the generation of a cloud of cold and dense gas beneath a forest canopy introduces a need to examine the processes facilitating exchange with the air above the canopy. It is the concentrations in this above-canopy air that are used to initialize many dispersion models and these concentrations are lower than would be expected if no canopy were present. In the lack of direct experimental studies of trans-canopy dilution in a forested environment, results from a variety of related experiments are used here to illustrate the complexity of the problem and to derive a first-order assessment of the extent of expected dilution. The focus is on the dense gas clouds following accidents involving liquid chlorine, ammonia and carbon dioxide. It is concluded that the dilution would result in above-canopy concentrations between 2% to 50% of average sub-canopy levels, depending on site-specific circumstances. It is also concluded that the relative importance of the various contributing processes is such that detailed incorporation of them in dispersion models would constitute an unjustified complexity unless definitive experimental observations are available. Instead, the consequences of the issues now considered might best be accommodated by reducing predicted downwind concentrations by a factor of about ten with a corresponding extension of the duration of the dispersion event.

Keywords: canopy; dilution; forests; intermittency; source term

Highlights

-- Dispersion model source-term concentrations can be less than subcanopy by a factor ≈ 10.

-- Episodic sweeps, ejections or bursts of turbulence facilitate trans-canopy exchange.

-- Canopy transparency and PBL influences appear to be major contributing factors.

-- Studies of trans-canopy exchange of gases are rare; research opportunities abound.

Introduction

Transportation of hazardous materials imposes acknowledged risks, confronting all of society with its unavoidable necessity. Accidents involving the transport of liquified chlorine (Cl_2) and ammonia (NH_3) are a societal reality affecting the eastern USA, which is largely forested. Accidental spills of liquid carbon dioxide (CO_2) have become a concern to the oil and gas industry. Many models have been constructed for predicting which areas are likely to be most affected by an accidental release of such chemicals, occurring at an unspecified time, at an unpredictable location, and in unexpected circumstances (Hanna et al., 2008). In practice, these models develop predictions of concentrations at downwind locations by assessing the effective sources and then building upon understanding of the many contributing processes of dispersion, deposition and chemical reaction. Comparisons among the predictions of such models generally indicate fairly good agreement, provided variations in wind direction (i.e. in the plume streamline) are accounted for. Otherwise, a major source of uncertainty relates to the specification of wind direction and the effects of meander.

Hanna et al. (2008) compare results from several models used to address the consequences of three accidental releases of liquid Cl_2 from railcar accidents. An overprediction of downwind concentrations is explained in terms of near-field deposition and chemistry. Here, an additional consideration is examined — the retention of a cold, dense gas within a sub-canopy airspace and its sporadic transfer to the air above the canopy. This will likely result in a reduction in the effective source term used in dispersion model initialization. Some models include consideration of this process but it is the intent here to focus on those that do not and to estimate how source terms must be modified, regardless of the specific model of interest.

There is a long history of field studies of liquid NH_3 spilled to specific areas. A series of experiments was conducted at the Nevada Test Site in 1983 ("Desert Tortoise," see Goldwire et al., 1985). A subsequent series of studies was conducted near Bordeaux in France in 1996 and 1997 (Bouet et al., 2004). The recent Jack Rabbit-I field studies (Fox and Storwold, 2011; Hanna et al., 2012) examined the consequences of early morning spills of liquid NH_3 and Cl_2 over the playa of the Dugway Proving Ground in Utah. Many similar studies of the gas cloud following release of liquid CO_2 have been conducted in Europe (Jamoisa et al., 2014). All known terrestrial experiments of this kind have been conducted over poorly vegetated or arid land. There are no known experiments that address the consequences of accidents resulting in a dense gas cloud beneath a forest canopy.

The need to consider how the presence of vegetation affects the dispersion of dense gases beneath a forest canopy came to a head with fears of the use of phosgene and mustard gas to impede the advance of allied forces through the jungles of the Malay Peninsula in World War II. An experimental program (led by Frank Pasquill) conducted in northern Queensland (Australia) served largely to allay fears. In practice, the released gas tended to linger beneath the vegetation canopy, slowly diffusing into the air above the canopy and spreading so that retreating forces were potentially at as much risk as advancing. This result exemplifies the characteristic behavior of present concern: the retention of gases within layers of the atmosphere that are isolated from the turbulence of the free atmosphere above by the presence of a layer of foliage and the consequent dilution associated with the transfer of the gas through the canopy. (Details of this activity can be found in the archives of the Australian War Museum in Canberra.)

Here, in the lack of direct experimental evidence to guide relevant dispersion modeling, the processes contributing to the exchange of air across a forest canopy and the consequent rate of transfer of a dense gas of subcanopy origin will be explored using observations made in other circumstances. The intent is not to identify ways in which dispersion models might be modified to take these considerations into account — such awaits the completion of an experimental program to refine understanding. Instead, the present purpose is to quantify the consequences of the main contributing processes using results from field studies conducted in other applications and to integrate their likely effects into an initial estimate of dense-gas trans-canopy dilution. This dilution factor could then be applied, with appropriate caution, to the concentration predictions of any dispersion model that does not simulate the processes now considered.

The Conceptual Scenario

Hanna et al. (2008) discuss the use of six dense gas dispersion models to address liquid chlorine releases following three railcar accidents. Two of these events (Mcdona, Texas, and Festus, Missouri) were in industrial areas with clumps or beltlines of trees. The third location (Graniteville, South Carolina) was more urban but with scattered trees and bordered by thin forest. Most of the six dispersion models assumed classical formulations of the surface boundary layer (SBL), employing the familiar micrometeorological roughness length and displacement height to describe the nature of the surface, with turbulence formulated correspondingly. Some dispersion models are addressing the case of a densely vegetated surface. For example, the SCIPUFF model of Sykes et al. (2007) uses the forest sub-canopy wind profile of Cionco (1985) as a foundation.

CO_2 is also transported as pressurized liquid (primarily in industrial situations), flashing to cold gas upon sudden release. Many relevant field studies have been conducted (e.g., Jamoisa et al., 2014) and the Pfast model (Witlox et al., 2014) has been developed to address the consequences of CO_2 accidents. In the case of NH_3, a dense gas cloud can arise as the initial liquid flashes to gas and then migrates according to terrain and the imposed wind field.

The considerations to follow are largely independent of the chemical species involved. The discussion relates to the way in which a cold, dense gas contained within the air space below the canopy can be transferred to the atmosphere above the canopy when the layer of foliage and the density gradient imposed by the presence of the cold, dense gas limit turbulent diffusion. In this case, classical micrometeorological approaches are of questionable relevance, since they assume exchange by a spectrum of eddies that no longer exists. Exchange across a vegetated canopy will still be somewhat diffusive but enhanced by sporadic events — “emissions” and “sweeps” as described by Shaw et al. (1989) and Katul et al. (1997). These events are the result of processes not necessarily of local origin, being variously attributed to SBL processes such as the convergence of katabatic flows (as observed in complex terrain research: e.g., Barthlott et al., 2010) or to large-scale planetary boundary layer (PBL) activities (Gao et al., 1989; Yagüe et al., 2017).

If transfer from the air beneath the canopy into the atmosphere above were a steady process, then subcanopy gas concentrations would first be expected to approach a limiting profile, probably approximating an exponential with height, but obviously varying as prevailing conditions change. After starting at a high value, concentrations at some specific height above the crown would also decrease as time progresses, again probably exponentially, to finally return to the background level of zero. (The likelihood of resuspension of sequestered chemical is presently ignored.) If there were no confounding factors, then estimates of the rate at which gas is lost to the air above the canopy could be derived using standard micrometeorological Fickian diffusion. However, in all situations of practical relevance exchange across the canopy is constrained by stability and by the vegetation.

In the discussion to follow, a first (extreme) consideration will be the regime in which turbulent mixing is a dominating factor, such as when concentrations are low enough that the density departures they impose will not be of consequence. In such a situation and if the canopy is sufficiently transparent, classical forest meteorological results may well be appropriate. In such circumstances, expectations of the exchange of dense gases present in low concentrations from below the canopy to above it might be estimated using a description of the wind and turbulence profiles through the canopy and extending to the forest floor as depicted, for example, by Cionco (1985). This case will be examined using observations made at a research tower erected within a mixed deciduous forest representative of the eastern U.S.A. This tower is located on Chestnut Ridge, in Oak Ridge, Tennessee (35.9311 N; 84.3323 W) and is referred to as the CHESS facility.

A second and more likely situation in which forest/atmosphere exchange is enhanced by sporadic "sweeps" and "ejections" will also be explored using measurements made at the same research tower. Such packets of air moving through a forest sub-canopy will not exhaust the air-space of its dense gas content. Rather, they will mix with the polluted air and carry some of its contents with them as they pass through the canopy. The extent to which such intermittent flows through a forest canopy scour dense gas from the sub-canopy airspace cannot be quantified, as yet, by actual measurements made in a forest environment. However, field results in other situations where intermittent excursions affect pools of gas constrained by vegetation and stability regimes at the surface provide guidance. Here, observations made of the scouring of CO_2 accumulated at night in comparatively low concentrations above a maize crop will be used to quantify the extent to which intermittent scouring removes gas from a surface gas pool. The mechanisms affecting the scouring of the maize CO_2 pool are discussed here as an analogue to the forest case — in both situations the scouring will penetrate into the layer of gas as far as the prevailing density gradient will permit. It is admitted that this is an assumption (among many) requiring experimental investigation.

The circumstances of an accident involving a release of NH_3 are substantially different, in that NH_3 is lighter than air whereas Cl_2 and CO_2 are not. Nevertheless, pooling of NH_3 resulting from strong local stratification has been observed, similar to the CO_2/maize situation. An example is the daytime accumulation of NH_3 emitted by swamp biota, when the swamp surface is considerably cooler than the incoming air (Lichiheb et al., 2021). In the case of a liquid NH_3 release, the resulting gas would quickly interact with atmospheric water vapor and aerosols, so that the visible "gas cloud" would likely be dominated by particles.

It is assumed that dispersion models will be initiated by time-averaged concentrations (χ_a) at height z_a (often 10 m above the displacement height d, sometimes 10 m above the canopy top — the crown) or alternatively by fluxes of gas at that height. Figure 1 is a schematic representation of the situation of current relevance, based on the big-leaf approach (Monteith, 1981). The pathways illustrated are designed to align with the discussion above and with the two-layer resistance scheme for bi-directional NH_3 exchange between vegetation and the atmosphere presented by Sutton et al. (2013). Spatial averages of controlling properties are represented by resistances accounting for (a) the deposition of the chemical of interest to foliage in the subcanopy airspace (R_f), (b) the transfer of gas through the canopy and up to the height of interest (z_a) where dispersion models are initialized (R_{ac}), and (c) the atmospheric coupling between height z_a and the greater height where concentration is zero (R_{aloft}). Concentrations (χ) within the canopy are assumed to, at first, increase with time as evaporation continues at the surface if a liquid is present and to eventually adopt a profile controlled by the many fluxes and intermittent exchanges occurring. After this initial phase, or if no liquid is present, concentrations decrease with time at a rate defining the storage capacity C in Fig. 1. The influence of sweeps and ejections (as presently interpreted and as identified in the diagram) is illustrated by a leakage resistance R_l, introduced intermittently via a switch. The conceptual association of these various resistances remains open to debate. Note that this schematic is greatly simplified. It is not intended to define a numerical simulation of the way in which the various considerations interface, but to guide the assembly of them into conclusions relevant to a forest like that found in the eastern USA.

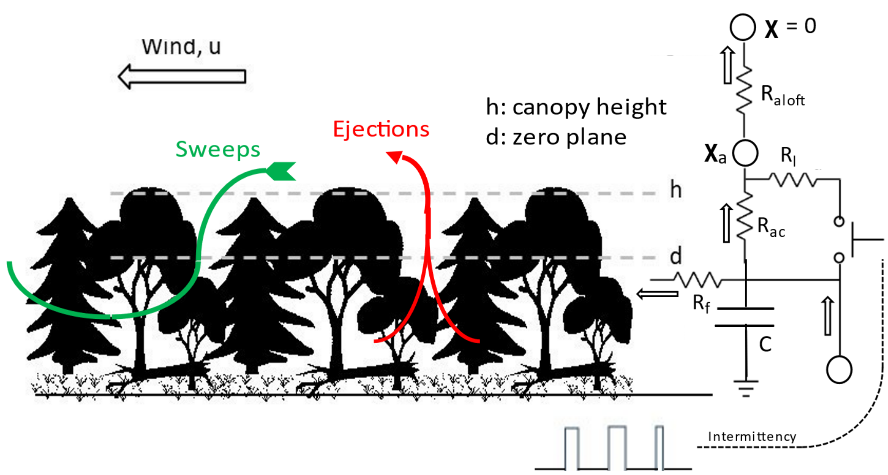

Fig. 1. Illustrating a simplified electrical analog describing the emission through a vegetated canopy of a trace gas pooled near the ground surface. Accumulation within the subcanopy airspace is represented by a capacity C.

Figure 1 is intended to illustrate two separate (and conceptually extreme) mechanisms by which gases entrapped within the sub-canopy air space can migrate into the atmosphere above. The first diffusive process operates continuously, according to familiar micrometeorology, with resistances R_f, R_{ac}, and R_{aloft} determining the association between the sub-canopy concentration (χ_c) and the concentration above the canopy (χ_a) at the height (z_a). The second pathway is associated with sporadic exchanges associated with sweeps and ejections, outside the realm of familiar Fickian diffusion theory. This pathway involves a new resistance R_l switched on and off intermittently. Note that the present visualization of sweeps and ejections departs from that of much recent literature. Whereas many examinations of the phenomenon (e.g., Finnigan et al., 2009) focus on fine-scale aspects of the processes involved, the present analysis considers no more than the large-scale processes revealed by measurements made every minute.

The resistance R_f describing transfer to foliage (or to whatever surfaces are available) necessarily involves the high-order chemical reactions quantified by Spicer et al. (2021) in their laboratory examination of a range of different plant species exposed to high concentrations of chlorine. These chemical processes necessarily involve reactions of the first and second order. That is, when concentrations are low the deposition will be like that of familiar dry deposition expressions with the flux being proportional to the concentration χ_a in air.

A Diffusive Baseline

Observations used here and in the section to follow were obtained using the CHESS tower, providing 10 Hz measurements of velocity and temperature at 5 m intervals from the forest floor up to a height of 60 m, recorded automatically as averages, variances, and covariances every 15 min. Observations made at two intermediate heights (22.5 m and 27.5 m) were also archived for a short period, to improve depiction through the forest canopy and across the plane of zero displacement of the logarithmic wind profile. One-minute temperature averages were archived separately.

The CHESS tower is in a mixed deciduous forest, mainly tulip poplar and oak, typical of much of the southern Appalachian region. Seasonal changes are substantial, primarily due to leaf loss as depicted in Fig. 2. The illustrated leaf area index observations were collected over an 8 yr period at a site about 2 km distant from CHESS – the Walker Branch Watershed (see Hutchison et al., 1986). There is no reason to suspect that the trees populating the CHESS site will display leafing characteristics different from those of the Walker Branch. In the present context, details of the obvious seasonal behavior indicates that the resistance to exchange between the air space beneath tree-top level and the wind field above would be greater in summer than in winter.

Classical models of forest canopy wind fields often derive from studies of managed forests and orchards (see Cionco, 1985). In the case of the CHESS location, the velocity profiles are often poorly quantified — within the subcanopy airspace the reported velocity components are frequently not greatly different from the reported standard deviations within the 15 min ensembles (as also reported by Su et al., 2019). However, when considered from the perspective of the averaged wind speed (as measured by three-cup anemometers) a more consistent profile arises. It is not clear how much classical forest canopy wind profiles are affected by consideration of speed rather than velocity. In any case, turbulent transfer of a trace gas through the canopy in question here will be determined by the level of turbulence beneath and through the canopy. The conventional measure of relevance is the turbulent kinetic energy (TKE).

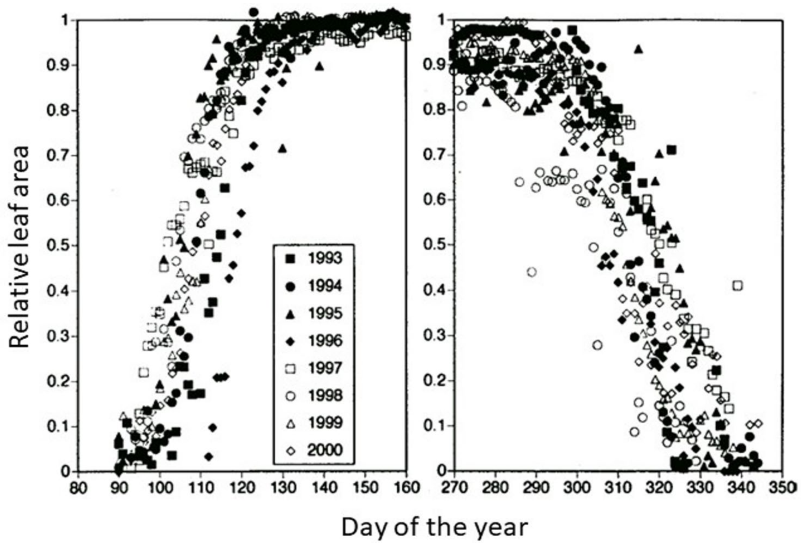

Fig. 2. Eight years of leaf area measurements made at the Walker Branch Watershed field site in Oak Ridge. The forest surrounding the Chestnut Ridge research tower is a mixed deciduous forest with a dominance of tulip poplar and oak, reaching to about 30 m height. These measurements were made by the team led by Boyd Hutchison of the NOAA Air Resources Laboratory (q.v. Hutchison et al., 1986).

Figure 3 presents observations of TKE (defined here as $\sigma_u^2 + \sigma_v^2 + \sigma_w^2$, where u, v and w are the conventional velocity components) made at the CHESS location. The observations indicate that regardless of canopy leafiness the TKE values near the ground tend to be about an order of magnitude less than those characterizing the SBL above the canopy. This is a well–understood characteristic of a forest environment (q.v. Su et al., 2019). The TKE decrease from levels seen above the canopy occurs rapidly through the canopy, comparatively slowly for the leafless case and more precipitously for the leafed. Note that the dawn profiles depart from this generalization — a matter inviting additional investigation. The observations identify the physical thickness of the mechanically obstructing layer — from 20 to 30 m above ground level for the fully leafed case at the time of these measurements. From the perspective of the diffusion of gases present in trace concentrations in the subcanopy airspace, it is levels of TKE observed within the canopy that determine the ability of the gas to diffuse through this barrier.

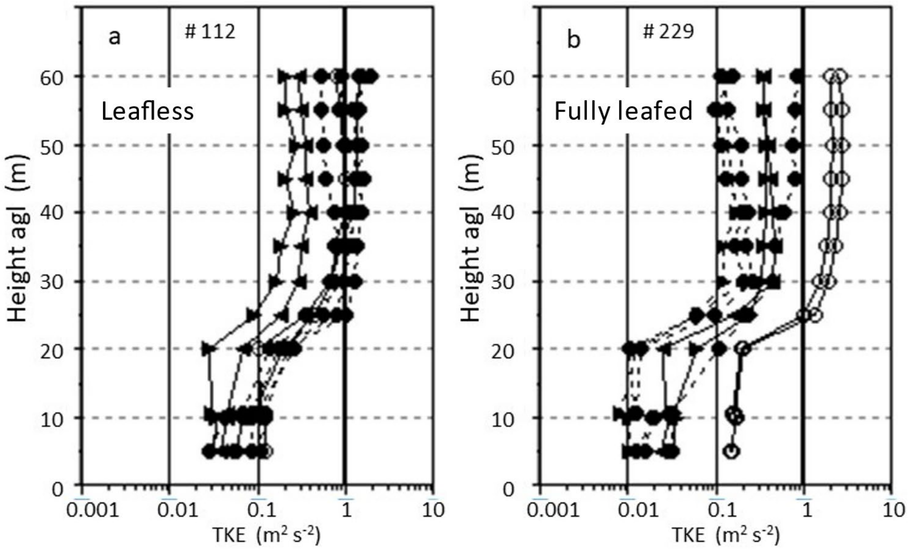

Figure 3. An example of the spread of one-hour TKE profiles observed at the CHESS site in Tennessee. Profiles observed at 3 h intervals starting at midnight are shown. (a) A sample of observations obtained during a 24 h period before budbreak in 2016 (day number as indicated), (b) during a day when the canopy was fully leafed. Observations made during the night are plotted using solid black circles; in daytime, open circles. Profiles measured near the dawn and dusk transitions are indicated by triangles: ► for sunrise, ◄ for sunset.

The TKE values indicated in Figure 3 permit estimation of a time scale (T_c) associated with the diffusive exchange of a trace sub-canopy gas. Assuming that 10% of the observed TKE is associated with vertical diffusion (i.e., that $\sigma_w^2 \approx$ TKE/10), then the four sub-canopy averages (night, day; leafed, leafless) derived from the plots of Figure 3 would yield four different estimates of the time constant associated with depletion by turbulent mixing of some gas accumulated in the sub-canopy airspace. For the daytime case of Fig. 3a (leafless), the average sub-canopy value of TKE (≈ 0.1 m^2 s^{-2}) would indicate a time scale $T_c \approx 5$ min (assuming target height is 30 m and estimating T_c as 30/0.1 s); for the nighttime case, the corresponding TKE value would be about 0.04 m^2 s^{-2}, leading to $T_c \approx 8$ min. The situation for a fully leafed canopy (Fig. 3b) is less well determined, but the results indicate values of $T_c \approx 3.5$ min for daytime and ≈ 12 min for night. Note that these approximations apply to a situation in which there is no cold, dense gas present and the sub-canopy air is not warmer or cooler than the air above. In the case of a strongly stratified airspace with a dense, cold gas occupying its lower levels, the present considerations would be inappropriate and longer residence times would be expected.

When diffusive exchange of sub-canopy trace gas dominates, with negligible consequence of episodic exchange events, the resistance scheme shown schematically in Fig. 1 can serve as a foundation on which to estimate the ratio χ_a/χ_c. The resistances R_f, R_{ac} and R_{aloft} are then the controlling properties. For the moment, disregard R_f. At night, R_{aloft} can be estimated from consideration of the velocity difference between the upper level of relevance and the level z_a where χ_a is to be determined. If a low-level jet is present, then available data indicate that this upper-level wind speed would typically be in the range 10 to 12 m s^{-1}. As an average of conditions prevailing, the momentum flux would be expected to be constant with height below this level (otherwise, air would be either accelerating or decelerating), whereupon the resistance R_{aloft} would be approximately $R_{aloft} = (u_{aloft} - u_a)/u_*^2$. The other contributing resistance of relevance is R_{ac}, representing the efficiency of exchange <u>*through*</u> the obstructing canopy. In the more familiar models of dry deposition, the aerodynamic resistance R_a is associated with exchange <u>*to*</u> the canopy, between the observing height z_a and the displacement height *d.* The displacement height is conveniently visualized as the effective level of action of the drag force acting on the canopy (Thom, 1971). Therefore, as a first approximation the total trans-canopy resistance to exchange is likely to be about $2 \cdot R_a$. Combining these considerations and introducing the canopy-induced wind "braking factor" as $B_f \equiv u_{aloft}/u_a$, it appears that

$$R_{aloft} \approx (B_f - 1)R_a \qquad (1)$$

and

$$R_{ac} \approx 2R_a. \qquad (2)$$

Hence,

$$\chi_a \approx \chi_c \cdot (B_f - 1)/(B_f + 1). \qquad (3)$$

Note that this heurism attempts no more than to quantify the trans-canopy resistance on the basis of the maximum wind speed observed in the wind profile and to use this to estimate the concentration observed at height z_a. It does not consider the role of convection, but it is tacitly assumed that the wind profile at heights above z_a is determined by the total diffusivity and that mass transfer at these heights is controlled similarly. The identification of the low-level jet as the relevant meteorological quantity no more than an aid to visualization of the scheme now presented. The same approach would apply regardless of time of day, without identifying a low-level jet as a causative factor, but with due awareness of the many inherent assumptions.

The braking factor B_f would be expected to vary according to the areal average friction coefficient and certainly with stability. In the case of the forest now considered, $B_f \approx 4$ would seem an appropriate expectation, in which case the ratio χ_a/χ_c would amount to a factor of 0.6. This applies to an extreme situation in which sub canopy air is well mixed and unaffected by either stratification imposed by the presence of a dense gas or sporadic turbulence incursions.

Sweeps and Ejections

In the situation of major interest here, a pool of liquid or a cloud of cold gas accumulated in the lowest few meters above ground level will impose a stratification that will curtail initiation of convection beneath the canopy and negate the diffusive mechanism considered above. The CHESS observations permit examination of the alternative sporadic processes that exchange air between the sub-canopy and the atmosphere aloft. It is now generally accepted that the exchange between a partially confined sub-canopy airspace and the free atmosphere above is predominantly via sweeps and ejections. These occur at sporadic but often fairly regular intervals as the structure of the canopy and the prevailing meteorological conditions ordain (q.v. Gardiner, 1994; Katul et al., 1997; 2006). The roles of sporadic gusts and incursions of turbulence through the canopy are represented by the switch and additional resistance in the schematic of Fig. 1.

The transfer of momentum is typically associated with gusts that carry bursts of momentum downwards from regions of higher wind speed aloft. A gust with sufficient turbulent energy will penetrate the canopy and initiate exchange of the sub canopy air with the atmosphere above. This process can occur in either stable or unstable stratification and corresponds to the sweep characteristic as will be discussed below. Further, in unstable conditions and in the absence of a stratification-inducing dense gas within the sub-canopy air space, insolation causes sub-canopy temperatures to rise until sufficient to initiate a convective episode and an ejection of sub-canopy air (q.v., the temperature ramps observed by Shaw et al., 1989 and detailed, for an Amazon forest, by Campanharo et al., 2008). In the case of a dense gas pool existing below the forest canopy, there is little chance that this process of convective initiation will be relevant. All of these intermittent processes will be influenced by topography, both local and regional, but here details of such causes and their site-specific repercussions will be ignored. Instead, the considerations to follow relate specifically to the terrain and forests of Eastern Tennessee — with gently rolling hills such that ejections could also be initiated by convergence of drainage flows at the forest floor (e.g., Chamecki et al., 2020). This matter has yet to be explored.

To examine this intermittency, temperature measurements have been used as an indicator of incursions of air from above into the subcanopy airspace. The observations used were obtained at 1-min intervals, using platinum resistance thermometers through and above the canopy. The 13 time series of CHESS 1-min temperature observations (one sequence for each height of measurement – 5, 10, 15, 20.25, 25, 27.5, 30, 35, 40, 45, 50, 55 & 60 m) have been assembled into the presentations of Figs. 4 and 5. The periods shown were selected at random from the record available, representing the month of August 2017 (when the forest was fully leafed). Temperature exceedances have been identified at each height of measurement, by identifying occasions when the time series of 1-min observations indicate temperatures differing from the average of the previous 10-min by more than two standard deviations. The resulting time series have then been combined to show the trajectories of exceedances as functions of height and time, covering the range of observations from 5 m to 60 m above ground level.

Figure 4 shows a sample period of night observations, in which positive temperature excursions indicate the infrequent occurrence of incursions of warmer air (from above) into the colder sub-canopy. The characteristic features of the incursions depicted in Fig. 4 are much as expected. Packets of turbulence propagate from above and reach the ground about 10 min after passing through the canopy. The frequency of occurrence is in the range 1 to 2 h^{-1}. If these incursions were to encounter a pool of cold gas, then some of the gas in the upper part of the dense gas cloud would be scavenged.

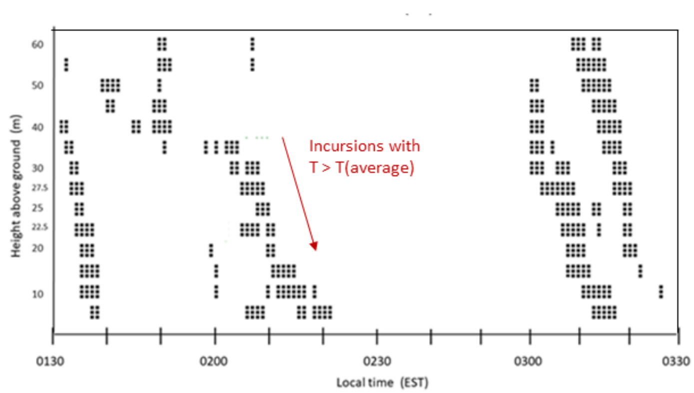

Fig. 4. An example of nocturnal incursions of warmer air ('sweeps') into and through the CHESS forest canopy. For each height of measurement, symbols are plotted whenever 1-min temperatures exceeded the average for the previous 10-min by more than two standard deviations.

In daytime, and in the absence of the cold dense gas now considered, positive temperature departures would indicate the consequences of convection generated at some level within the canopy airspace or some prevailing topographic consequence — an ejection and probably not relevant in the present cold, dense gas situation. More appropriately, a negative exceedance would relate to the replacement of the convectively-displaced air or (particularly in the current cold, dense gas situation) by an incursion from above due to other causes — probably associated with gusts migrating with the ambient wind and resulting in "sweeps." Figure 5 shows a sampling of the daytime occurrences of exceedances; when incursions from above carry colder air into the subcanopy. The illustration shows that the colder air incursions occur with about the same frequency as indicated in Fig. 4 — about 1 to 2 h^{-1}. However, in Fig. 4, the incursions propagate slowly downwards through the canopy. In Fig. 5, the downward sub-canopy movement is considerably faster. It is acknowledged that the selection of a 2σ criterion for identifying incursions introduces a limitation on their detection and identification, but examination of this would require more and better data than are presently available. Moreover, note that the present method of analysis does not permit detection of occurrences due to flow convergence.

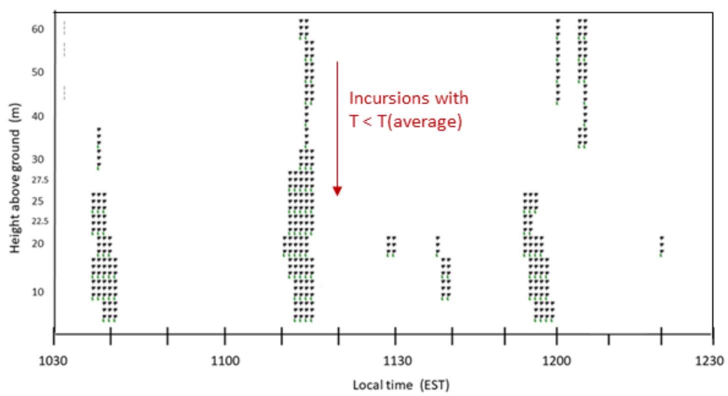

Fig. 5. An example of daytime incursions of colder air (sweeps) into and through the CHESS forest canopy. For each height of measurement, symbols are plotted whenever 1-min temperatures were below the average for the previous 10-min by more than two standard deviations.

Scouring by Sporadic Turbulence Incursions

Having estimated the frequency with which turbulence incursions occur in a forest environment typical of the eastern USA, and noting that this repetition frequency appears to be about the same for other vegetated areas (see Cionco and Ohmstede, 1992) and particularly for forested environments (e.g., in Europe — Yagüe et al., 2007), it remains to estimate the efficiency with which such episodes can remove the relevant dense gas. In the lack of direct experimental evidence, we turn instead to results obtained in a 2018 study of pooling of CO_2 above a maize canopy. CO_2 is continuously emitted from soil, at a rate depending on soil temperature, due to sub-surface biological activity. At night, CO_2 concentrations in air near the surface increase as soil emissions continue while exchange with the air above is limited if not curtailed due to nighttime stratification. Concentrations near the ground sometimes exceed 1000 ppm (c.f. a “normal” concentration of about 400 ppm), much lower than the concentrations of main interest in the context of a dense gas but uniquely revealing of intermittent scouring. Figure 7a illustrates a sequence of CO_2 diurnal cycles measured at 10-min intervals a short distance above the crown of a 2018 maize crop growing in Tennessee (about 15 km from the CHESS site, at 35.707ºN, 84.373ºW). The time sequence shown is for a period when the crop was mature — the second half of July. The canopy depth was approximately 2.8 m and CO_2 concentrations were measured at about 1.5 m above the crown. Similar behavior has been observed in all similar studies of CO_2 in and over a maize canopy, including in Africa (Hicks et al., 2015). It is clear that CO_2 pooling is an endemic phenomenon and that it is subject to periods of scouring occurring every night but with irregular periodicity and magnitude, much the same as was discussed above for the forest situation. Figures 7b and 7c are expanded to depict the sporadic occurrences in more detail and to show that they are not always as frequent. Most often, however, the plots shown indicate that each intermittent event causes a reduction in CO_2 concentration by about 10% (c.f. Zorzetto, et al., 2021). Elsewhere, these sporadic events were associated with PBL gravity waves and jets (Hicks et al., 2015) but there appear to be several possible causes (Chen et al., 2020) with elevated shear layers also being promoted as contributors (Dupont and Patton, 2012). Regardless of the specific mechanisms, examinations of turbulent intermittency have repeatedly identified the cause as being external to the SBL of conventional micrometeorology and more a consequence of mechanisms within the PBL (q.v. Mahrt, 1989).

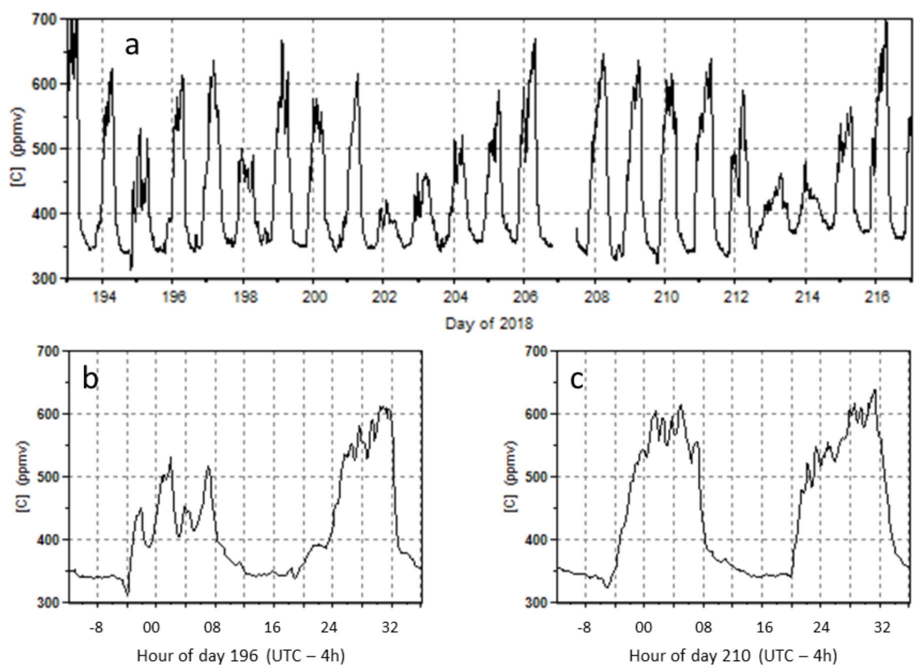

Fig. 7. Examples of observed diurnal cycles of CO_2 concentrations immediately above a maize crop in Loudon, Tennessee in 2018, showing (a) the ubiquity and variability of CO_2 nocturnal gas pooling, and (b, c) expanded examples of specific occasions indicating a loss of pooled gas by about 10% by most events.

In concept, the extent to which an intruding "sweep" will penetrate a layer of increasing density should not change greatly between the maize and the forest situations — in both cases the controlling factor is the density gradient that is encountered. However, it also appears likely that the density gradient of relevance would be much greater for the dense gas forest circumstance than for the maize, and hence the forest effective scouring efficiency would be less than the 10% now estimated. Regardless of this, the deliberations to follow assume a scouring efficiency of 10% per event. The uncertainties involved are reluctantly accepted, there being no relevant forest observations.

Trans-Canopy Deposition

So far, there has been no consideration of the uptake of gas as the polluted air passes through a vegetative canopy. In consideration of the processes described here, the chemical affinity of the gas with the plant species of relevance must be considered if a more defensible conceptual construct is desired. In practice, the passage of gas through the canopy above a partially confined airspace will entail a deposition rate that is estimable (provided concentrations are adequately specified).

In practice, the presence of Cl_2 in high concentrations would cause stomata to close and the deposition of relevance must then be limited to cuticular exchange. The extensive examinations of plant responses to Cl_2 exposure conducted in the 1940s and 1950s (e.g., McCallan and Weedon, 1940; Thornton and Setterstrom, 1940) indicate a typical necrotic dose of about 200 ppm hr. This value can be used as an entry point for detailed consideration of the reduction of concentrations affecting dispersion model initialization due to trans-canopy deposition. Recent laboratory studies with Cl_2 have revealed that higher-order chemical reactions must be considered in the vicinity release when concentrations are high (Spicer et al., 2021).

For NH_3, the matter is complicated by the biological prevalence of NH_3. Depending on its nitrogen status, vegetation can act either as a sink or a source of NH_3 (Sutton et al., 1995, 2013). The magnitude and direction of the exchange depends on the canopy compensation point (χ_{comp}), defined as the atmospheric NH_3 concentration for which the flux between the canopy surface and the atmosphere changes from emission to deposition (or vice versa; Farquhar et al., 1980). Two exchange pathways at the plant level determine χ_{comp}. First, due to its high solubility in water and polarity, NH_3 can rapidly deposit to a wet leaf cuticle. Second, NH_3 can enter open stomata and be captured by mesophyllic tissue. This stomatal pathway is bi-directional and depends on the NH_3 concentration difference between the surrounding air and the sub-stomatal cavity, considered in terms of χ_{comp}. The stomatal compensation point can be approximated using knowledge of the contributing biological processes and the nitrogen status of the plant tissue (Hill et al., 2002; Massad et al., 2010). In an accidental release of NH_3 affecting forest vegetation, an uncommonly large χ_{comp} must be expected. The large canopy compensation point will lower the rate of deposition at different levels within the canopy and lead to higher emissions at subsequent times.

The biological factors controlling exchange of CO_2 between the air and foliage have been well studied and widely reported, although with a dominance of attention to aspects related to climate change and not extending to the high concentration regimes of current relevance (e.g., Law et al., 2002). If the matter of CO_2 releases into a forest environment is seen to be important, then relevant field experiments would be advised.

The overall adjustment required in estimating χ_a after taking deposition into account is difficult to estimate. Certainly, an estimate based on studies of near-field Cl_2 (Spicer, 2021) will be different from expectations for NH_3. For the Cl_2 case, exposure of foliage to Cl_2 will likely cause immediate stomatal closure, with recovery being much slower after the passage of a high-concentration event. However, Spicer et al. (2021) report that "trees were found to either be relatively ineffective at removing chlorine . . . . or intermittently effective . . . ." In comparison, NH_3 will not cause stomatal closure until the dose exceeds some limit that is not well known. Once deposited and retained in plant tissue, NH_3 can be returned into the air after air concentrations drop to beneath a compensation point that depends on how much NH_3 was previously deposited.

Without relevant field observations, the effective retention during air transfer through a forest canopy cannot be quantified, but on the basis of studies conducted in other circumstances it appears likely that the deposition of the gas (Cl_2, CO_2 or NH_3) to canopy foliage will not be a major contribution to trans-canopy dilution. A value of 1% removal during the leafed season seems reasonable, lower (of course) if trees are leafless.

Discussion

The baseline diffusive transfer of sub-canopy air into the wind stream aloft was estimated above (Eq. 2 et seq.) to impose a reduction in concentrations to about 60% of average sub-canopy levels. This estimate relates to a situation in which the sub-canopy is unaffected by the presence of a dense gas or temperature regime that would impose a stability constraint on mixing. In the situation of current interest, the presence of high concentrations of a cold gas cloud within the sub-canopy airspace would necessarily impose a stability regime that would eliminate this background reference level from consideration. It is apparent, however, that the influence of this imposed stratification would result in more dilution than the $\chi_a/\chi_c \approx$ 60% estimated reference level.

When the canopy is fully leafed and the sub-canopy contains a cloud of cold dense gas arising from settling from a liquid jet emission from a ruptured container or from an evaporating liquid spill, the trans-canopy exchange of the gas would mainly be due to episodic scouring of gas at the top of the gas cloud. Identifying such an episode with any particular location and time does not seem amenable to deterministic analysis. Although it is common to associate a fairly regular periodicity with the phenomenon, the matter is one of statistics and chaos as well as the prevailing synoptics. The maize crop observations of Figure 7 indicate that typical sweeps scour about 10% of the available gas cloud. If the same applies to the case of the CHESS site, where the events appear to occur at a frequency of about 2 h^{-1}, about 20% of the sub-canopy gas cloud would be removed every hour. The residence time of the cloud beneath the sub-canopy would then be of the order of hours, much longer than that derived for the diffusion-influenced reference situation considered above (many minutes).

The material scoured from the sub-canopy gas field would enter into the atmosphere above the crown and be mixed with inflowing air. With a time scale of about 5 min associated with the duration of every event (as indicated in Fig. 5 and discussed in the relevant text) a further dilution of one part in six would seem an appropriate measure. Overall, it appears that the likely effective dilution corresponding to the height z_a where dispersion models are initialized would be by about a factor of about sixty, computed as the event scouring efficiency (10%) times the proportion of the time relevant (5 min every 30 min = 1/6).

In the end, the present deliberations indicate a likely dilution to be 1% to 2% of sub-canopy concentrations when the gas is present at high concentrations and if the deciduous canopy is fully leafed. This proportion will increase as sub-canopy concentrations decrease, eventually approaching a level that could be as high as 60% (sweeps and synoptics permitting). These estimates represent two extreme cases — first relating to when the sub-canopy gas field is pooled and concentrated and second to when the sub-canopy is well mixed and the gas of interest can be viewed as a passive trace contaminant. Without additional site-specific knowledge and more detailed understanding, the most likely concentration appropriate for the initiation of dispersion models at all stages of the event and its aftermath would best be estimated as about 10% of the concentration representative of the subcanopy airspace, this being the geometric mean of the two extremes.

Conclusions

The basic question addressed here relates to the way in which concentrations of cold, dense gas contained within the sub-canopy airspace of a forest must be adjusted to provide a good starting point for dispersion calculations. It has not been the intent to answer the basic question in a robust and defensible manner. Rather, the goal has been to demonstrate a way in which the processes of relevance can be conceptualized and their interactions considered. A necessary consequence is that the enumerations of trans-canopy dilution developed here must not be taken as more than heuristic estimates, awaiting field examination.

Understanding of the processes involved in the behaviors now being considered is evolving at this time. The modified electrical analog summarized in Fig. 1 appears to offer a way to address the effects of intermittency once some basic characteristics of the mechanism are quantified and accounted for (such as how a sub-canopy dense gas cloud is created and maintained, and how it is depleted by diffusion and deposition once atmospheric stability considerations are introduced). Alternatively, consideration of intermittent incursions provides a more mechanistic entry point into the same puzzle. After combining results derived from considerations of the separate contributing processes, it is concluded that a good first estimate of the dilution associated with the presence of a forest canopy would likely be by about a factor of ten, on average. This estimate would necessarily change according to (for example) season, forest species constituency, and the characteristics of the initial injection of gas (or liquid) into the forest. The wide variability of each of the contributing processes is such that little confidence can be associated with this average and no embellishment of existing models can be recommended. Instead, it seems most appropriate for the present dilution estimate to be considered as a factor modifying the outputs of existing dispersion models and with levels of prediction uncertainty derived accordingly.

There are no known field observations of the transfer of subcanopy dense gas into the wind field above a forest. In the lack of such direct measurements, use has been made of results of recent field experiments addressing other issues, but all within the same topological domain in eastern Tennessee. The considerations presented here are based on opportunities presented by field programs in which the present authors participated. Similar opportunities are expected to arise, elsewhere. Analyses enabled by such assemblies of experimental results will probably continue to provide guidance to the dispersion modeling community, in the context of forests, until field experiments permit description and quantification of trans-canopy dilution.

The complexity of the processes contributing to trans-canopy dilution is such that detailed description suitable for inclusion in specific models is not recommended. Since the same dilution would affect the application of any dispersion model, it is recommended instead that a single quantity, the canopy concentration dilution ratio χ_a/χ_c (or some equivalent) be accepted as an empirical factor to be determined by examination of many forest situations and described in terms of a few quantifying variables such as season, topography, forest species and chemical species. It is unlikely that it will be feasible to conduct repeated field studies involving releases of chemicals like Cl_2 or NH_3 in real-world forested surroundings. Instead, it is proposed that field experiments be initiated to explore the processes discussed here and the ways in which they interact. Experiments using injections of CO_2 below the canopies of existing forest meteorology locations (already instrumented and with local expert researchers) would offer a cost-effective strategy with high likelihood of success.

To capitalize on the ubiquitous nature of CO_2 emanation from soil, it is recommended that existing forest tower installations record observations of the standard meteorological variables and of CO_2 concentrations at 1 min frequency or more frequently. This would enable contributing sporadic events to be detected and evaluated as has been demonstrated here using the CHESS observations. CO_2 concentrations are easily monitored (potentially using modern solid-state sensors). Sensors should be deployed as situations permit — beneath, through, and above the canopy.

Acknowledgements

Comments and suggestions provided by anonymous reviewers are truly appreciated. The CHESS experimental site in Oak Ridge is operated by NOAA/ARL. The CO_2 experimentation in Loudon was supported by the University of Tennessee. The principal author serves as a contractor to NOAA/ARL. The scientific results and conclusions as well as any views and opinions expressed herein are those of the authors and do not reflect the views of any of the sponsoring agencies.

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Statement of novelty

The emergency response community has been struggling to improve models of dense gas dispersion after a leak of stored liquid. Current models overlook the complex role of a vegetation canopy, should one exist. In the lack of direct experimental evidence, the present analysis makes use of recent forest meteorology observations and details of CO_2 pooling over crops to derive an estimate of the dilution expected when subcanopy Cl_2 and/or NH_3 concentrations are used to initiate conventional dispersion models. The estimated dilution amounts to about an order of magnitude.

Summary of Co-Author Contributions

Nebila Lichiheb — Details related to NH_3 dispersion and deposition.
Neal S. Eash — Design and support for the CO_2 experimentation.
Joel N. Oetting — Field experimentation and data analysis.

Environmental Implication

This work pertains to the provision of dispersion forecasts to protect the public and facilitate emergency response in the event of an accidental release of liquid chlorine, carbon dioxide or ammonia into a forested environment. Predictions of contemporary dispersion models are based on experiments conducted over barren terrain and are not directly relevant to conditions encountered in eastern North America (which is largely forested). It is estimated that current models (neglecting the role of vegetation) overestimate risk by an order of magnitude.

Highlights

-- Dispersion model source-term concentrations can be less than subcanopy by a factor ≈ 10.

-- Episodic sweeps, ejections or bursts of turbulence facilitate trans-canopy exchange.

-- Canopy transparency and PBL influences appear to be major contributing factors.

-- Studies of trans-canopy exchange of gases are rare; research opportunities abound.