Text S-1: Near-source Gaussian plume slopes.

If we integrate the Gaussian plume solution (2) with respect to the crosswind-direction *y*, we obtain

$$\frac{\overline{c(x,z)}}{S} = \frac{1}{\sigma_z(x)} \exp\left(\frac{-z^2}{2\sigma_z^2(x)} \right) \tag{S1.1}$$

with $S \equiv \sqrt{\frac{2}{\pi}}\left(\frac{Q}{U}\right)$. If we treat $\overline{c(x,z)}$ as a constant while differentiating (S1.1) with respect to *x*, we get an implicit equation for the slope dz/dx of that concentration surface with respect to downwind distance. After rearranging terms we have:

$$\frac{dz}{dx} = \left[\frac{z}{\sigma_z} - \frac{\sigma_z}{z} \right] \frac{d\sigma_z}{dx} \tag{S1.2}$$

This can be simplified if we choose a concentration surface much more than one standard deviation away from the maximum concentration (e.g., one that could be designated the 'edge' of the plume); then $z >> \sigma_z$, and the second term in brackets can be neglected. Since $\sigma_Z^2 = x^2\sigma_W^2 / U^2$ in the near-source limit, the slope of the edge of the plume should have the approximate equation:

$$\frac{dz}{dx} \approx \left[\frac{Uz}{x\sigma_W} \right] \frac{\sigma_W}{U} = \frac{z}{x} \tag{S1.3}$$

The solution to this is $z = Cx$, implying that this concentration surface has a constant slope in the vicinity of the source. (Note that near a surface point source both *z* and *x* on a concentration isopleth must approach zero.) However, if we applied the far-source expression $\sigma_Z^2 = 2xT_{LZ}\sigma_W^2 / U^2$ instead, we get:

$$\frac{dz}{dx} \approx \left[\frac{z}{\sqrt{\frac{2xT_L}{U}}\sigma_W} \right] \sqrt{\frac{2T_L}{U}} \frac{\sigma_W}{2\sqrt{x}} = \frac{z}{2x} \tag{S1.4}$$

whose solution is $z = Cx^{1/2}$, which has an infinite slope near the origin. Thus the integrated concentration surfaces ascend almost vertically near the source, implying immediate vertical mixing; with increasing distance the surfaces become nearly but never quite horizontal. By contrast, integrated concentration surfaces using the near-field expression should maintain a constant slope near the source, implying that for each concentration threshold, there is a finite downwind distance, increasing with height above the surface, before that threshold will be observed. The smaller rate of ascent of concentration contours implies higher near-surface integrated concentrations for the same downwind distance.

Text S-2: Legendre solution to convective PBL plume

For further quantitative analysis of the convective PBL we would like to have an expression that smoothly merges the near-field and far-field behavior of the Taylor theory. However, we also want into to take into account the observed fact that the vertical velocity variance is not constant with height in the daytime PBL, but has a mid-PBL maximum. We also want the solution to reflect the fact, unlike in the unbounded Gaussian plume model, the growth of the plume in the vertical does not proceed indefinitely; rather, the tracer is largely confined to be below the mixed layer height, z_i.

We now try to quantify some of these theoretical expressions based on statistics from the daytime convective PBL, and compare them to the modeled plumes. We assume that the Lagrangian timescale of the turbulence is given by $T_L = cz_i / w_*$, where c is a constant whose value varies somewhat in the literature but was estimated to be 0.4 by Dosio et al. (2005). For the vertical velocity variance we assume that $\sigma_W^2 = 4aw_*^2 \left(\frac{z}{z_i}\right)\left(1-\frac{z}{z_i}\right)$. This function reproduces reasonably well the observed mid-PBL maximum of vertical velocity variance in the convective PBL; the value of $a = \sigma_{W\max}^2 / w_*^2$ is approximately 0.4 (e.g., Stull 1988; Arya 1995). So for the vertical diffusion coefficient we assume:

$$K_z = \sigma_W^2 T_L f(\hat{x}) = 4acw_* z_i \left(\frac{z}{z_i}\right)\left(1-\frac{z}{z_i}\right) f(\hat{x}) \qquad \text{(S2.1)}$$

where $\hat{x} \equiv \frac{x}{UT_L} = \frac{w_* x}{cUz_i} = \frac{x}{cx_0}$. For a mesoscale PBL scheme we assume $f(\hat{x}) = 1$. However, for the LES we assume that:

$$f(\hat{x}) = 1 - e^{-\hat{x}} \qquad \text{(S2.2)}$$

This function is consistent with that of Taylor (1921) and reproduces both the short-time and long-time limits. Finally, we assume that $K_y = \sigma_V^2 T_L f(\hat{x}) = bcw_* z_i f(\hat{x})$, where $b \equiv \sigma_V^2 / w_*^2 \approx 0.2$ (e.g., Willis and Deardorff 1976; Moeng et al. 2007).

Given our prior meteorological assumptions, the steady-state plume from a constant surface point source for the differential equation

$$U\frac{\partial c}{\partial x}-\frac{\partial}{\partial y}\left(K_y\frac{\partial c}{\partial y}\right)-\frac{\partial}{\partial z}\left(K_z\frac{\partial c}{\partial z}\right)=Q\delta(x-0)\delta(y-0)\delta(z-0) \qquad \text{(S2.3)}$$

can be found by first performing a Fourier transform over *y*, which converts the *y* derivative into an algebraic expression that is converted back to *y* at the end with the inverse transform. The resultant equation is separable for *x* and *z*; the *x*-equation has an exponential as a solution. With boundary conditions of zero vertical flux at the lower boundary and at z_i,,the *z*-equation has eigenfunctions that involve Legendre polynomials (e.g., Nieuwstadt 1980; Otte and Wyngaard 1996). The unique regular analytical solution is given by:

$$c(\hat{x},y,\hat{z})=\frac{Q}{Uz_i}\frac{1}{\sqrt{4\pi bc^2z_i^2q(\hat{x})}}\exp\left(\frac{-y^2}{4bc^2z_i^2q(\hat{x})}\right), \qquad \text{(S2.4)}$$
$$\times\sum_{n=0}^{\infty}(2n+1)P_n(1-2\hat{z})\exp(-4ac^2n(n+1)q(\hat{x}))$$

Here $\hat{z}\equiv z/z_i$, $P_n(\,)$ are the Legendre polynomials, and:

$$q(\hat{x})\equiv\int_0^{\hat{x}}f(x')dx' \qquad \text{(S2.5)}$$

For the "mesoscale" / far-source model, $q(\hat{x})$ is just the normalized downwind distance, $\hat{x}$, but for the "LES" / blended model, is given by the monotonic function $q(\hat{x})=\hat{x}-(1-e^{-\hat{x}})$.

The corresponding crosswise-integrated plume concentration is:

$$c_{\text{int}}(\hat{x},\hat{z})=\frac{Q}{Uz_i}\sum_{n=0}^{\infty}(2n+1)P_n(1-2\hat{z})\exp(-4ac^2n(n+1)q(\hat{x})) \qquad \text{(S2.6)}$$

From this it can be seen that for either far-source or blended plume, the crosswise-integrated concentration becomes well-mixed at concentration $Q/(U z_i)$ as $\hat{x}$ gets large.

Text S-3: Slopes of cross-plume integrated Legendre solutions near surface source

The cross-wind-integrated steady-state plume equation for a constant surface point source, given assumptions discussed in the text, is:

$$U\frac{\partial c_{\text{int}}}{\partial x} - \frac{\partial}{\partial z}\left(K_z \frac{\partial c_{\text{int}}}{\partial z}\right) = Q\delta(x-0)\delta(z-0) \tag{S3.1}$$

As stated in Supplementary Material Text S-2, the solution to this equation, given the vertical diffusivity:

$$K_z = \sigma_W^2 T_L f(\hat{x}) = 4acw_* z_i \left(\frac{z}{z_i}\right)\left(1-\frac{z}{z_i}\right) f(\hat{x}) \tag{S3.2}$$

and boundary conditions of regularity at $z = 0$ and $z = z_i$ is:

$$c_{\text{int}}(\hat{x},\hat{z}) = \frac{Q}{Uz_i}\sum_{n=0}^{\infty}(2n+1)P_n(1-2\hat{z})\exp(-4ac^2 n(n+1)q(\hat{x})) \tag{S3.3}$$

Here, $\hat{x} \equiv \dfrac{x}{cx_0}$, $\hat{z} \equiv z/z_i$, and

$$q(\hat{x}) \equiv \int_0^{\hat{x}} f(x')dx' \tag{S3.4}$$

The reader is referred to Supplementary Material Text S-2 and the main text for the meaning of other symbols.

We wish to find the slope of the integrated concentrations surfaces near the source and close to the ground. One way to do this is actually to consider a modified problem, consisting of (S3.1) with vertical diffusivity:

$$K_z = 4acw_* z_i \left(\frac{z}{z_i}\right) f(\hat{x}) \tag{S3.5}$$

The modified solution should be a good approximation to the original solution provided $\hat{z} = z/z_i << 1$ (i.e., the height is much less than the mixed layer height). Additionally, we can take the top boundary condition for the modified problem to be that the solution remains bounded as *z* approaches infinity, with the understanding that only the $\hat{z} << 1$ part of the solution will be physically meaningful. The primary advantage of considering the modified solution is that, unlike the original solution (S3.3), there is a single continuous solution (except at the origin) instead of a discrete set of eigensolutions, which makes it easier to find the slope of the total integrated concentration.

It can be shown that the solution to the modified problem (S3.1) and (S3.5) is:

$$c_{\text{int}}(\hat{x},\hat{z}) = \frac{Q}{UAq(\hat{x})x_0}\exp\left(-\frac{z_i\hat{z}}{Aq(\hat{x})x_0}\right) \tag{S3.6}$$

in which $A \equiv 4acw_* / U$. To find the slope of an isopleth of integrated concentration, we then take:

$$\frac{\partial \hat{z}}{\partial \hat{x}}\Big|_{c\,\mathrm{int}} = -\frac{\dfrac{\partial c_{\mathrm{int}}}{\partial \hat{x}}\Big|_{\hat{z}}}{\dfrac{\partial c_{\mathrm{int}}}{\partial \hat{z}}\Big|_{\hat{x}}} \tag{S3.7}$$

For the mesoscale case, $q(\hat{x}) = \hat{x}$. Substituting, we eventually get:

$$\frac{\partial \hat{z}}{\partial \hat{x}}\Big|_{c\,\mathrm{int}} = \frac{\hat{z}}{\hat{x}} - \frac{Ax_0}{z_i} \tag{S3.8}$$

So for the mesoscale case the slope of an isopleth is zero when $\hat{z} = Ax_0 \hat{x} / z_i$. It follows that the maximum vertical extent of all the mesoscale concentration surfaces falls on a line from the origin with constant slope. This behavior can be seen for example in the full analytical solution in the left panel of Figure 13, as well as the mesoscale WRF plots in Figures 11-12.

If we now take a point much closer to the origin horizontally than the location of the zero slope isopleth at a given height $\hat{z}$, then we may neglect the second term on the RHS of (S3.8). Treating this as a differential equation of $\hat{z}(\hat{x})$, representing the height of the isopleth near the origin, we solve to obtain $\hat{z} = C\hat{x}$. The concentration surfaces thus approach straight lines at the origin.

For the LES case, $q(\hat{x}) = \hat{x} - (1 - e^{-\hat{x}})$, and more involved algebra yields:

$$\frac{\partial \hat{z}}{\partial \hat{x}}\Big|_{c\,\mathrm{int}} = \left[\frac{\hat{z}}{\hat{x} - (1 - e^{-\hat{x}})} - \frac{Ax_0}{z_i}\right]\left[1 - e^{-\hat{x}}\right] \tag{S3.9}$$

If we now also assume that $\hat{x} << 1$, then we can approximate the exponential by a Taylor series truncation. To lowest order:

$$\frac{\partial \hat{z}}{\partial \hat{x}}\Big|_{c\,\mathrm{int}} \approx \left[\frac{2\hat{z}}{\hat{x}^2} - \frac{Ax_0}{z_i}\right]\hat{x} \tag{S3.10}$$

Since $\hat{x} << 1$, if we assume as in the mesoscale case $\hat{x} << z_i \hat{z} / Ax_0$, then the second term in the brackets of (S3.10) can be neglected. Solving the approximate differential equation now gives $\hat{z} = C\hat{x}^2$.

So in the LES case the isopleths near the origin are shaped like concave-up parabolas, as can be seen in the right panels of Figures 11-13. Most notably, this shows that all the isopleths of integrated concentration became tangent to the surface at the origin, leading to a vertical concentration gradient theoretically becoming infinite. While obviously the analytical model would break down before this point, this analysis does seem consistent with the very large near-surface concentrations apparent in the LES near the source.

Table S-1: Approximate height of lowest model layers above ground level.

Index of Model Layer from Ground	Approximate Height of Model Layers above Ground (m)	Approximate Height of Center of Model Layer above Ground (m)
1	0 – 13.8	6.9
2	13.8 – 31.4	22.6
3	31.4 – 53.6	42.5
4	53.6 – 79.6	66.6
5	79.6 – 110.4	95.0
6	110.4 – 144.8	127.6
7	144.8 – 184.4	164.6
8	184.4 – 227.4	205.9