The current review concentrates on two topics related to the calculation of fluxes by the gradient technique employed in the study.

Calculation of eddy diffusivity

By gradient technique the vertical turbulent flux can be represented by

$$F(z) = -K_z \frac{\partial \overline{C}}{\partial z}.$$

The vertical eddy diffusivity can be parameterised as $K_z = \tau \sigma_w{}^2$, where τ and σ_w represent the Lagrangian turbulent time scale and the standard deviation of vertical velocity, respectively.

The authors state that the time scale is the unit time scale (1 s). This cannot be the case in general because the Lagrangian turbulent time scale depends on the observation level as well as turbulent state of the atmosphere. Justification of using 1 sec as the relevant time scale is needed.

For example, the Lagrangian stochastic models employ frequently for the atmospheric surface layer (ASL) dispersion the time scale defined by $\tau = \frac{2\sigma_w^2}{C_0 \varepsilon}$ (e.g. Wilson and Sawford, 1996, Boundary-Layer Meteorol. 78, 191-2010, 1996), where C_0 stands for the Kolmogorov constant. By using the common ASL parameterisation for the dissipation rate of the turbulent kinetic energy $\varepsilon = \frac{u_*^3 \phi_\varepsilon}{k\, z}$, where u_* is the friction velocity, k is the von Karman constant (0.4), and ϕ_ε the stability function for ε (see e.g. Kaimal, J. C., and Finnigan, J. J., Atmospheric Boundary Layer Flows. Their Structure and Measurement, Oxford University Press, New York, 1994), and the common parameterisation for $\sigma_w = 1.25\, u_* \phi_w$, where ϕ_w is the stability function for σ_w, the expression for the Lagrangian time scale becomes $\tau = \frac{2\left(1.25^2\right)\phi_w^2 k\, z}{C_0 u_* \phi_\varepsilon}$. For neutral conditions it can be expressed as $\tau = \dfrac{2\left(1.25^2\right) k\, z}{C_0 u_*}$, showing that the time scale is proportional to height and inversely proportional to u_*.

The study involves measurement of turbulent statistics by the EC system, including the momentum, sensible and latent heat fluxes. Those measurements define the friction velocity u_* as well as the Obukhov length scale characterising the stability of the ASL. Further, the ASL similarity theory would allow to infer the eddy diffusivity via $K_z = \dfrac{k\, z\, u_*}{\phi_c}$, where ϕ_c is the stability function for scalar (heat), can be taken e.g. from Kaimal and Finnigan (1994). Under near-neutral conditions the stability function ϕ_c approaches unity and the eddy diffusivity is defined only by the observation level and the friction velocity (inferred from the measured momentum flux). This would be perhaps most direct way to obtain the eddy diffusivity from the available measurements. The authors could use this definition of the eddy diffusivity or at least verify their approach by comparing with this well established definition of K_z.

Averaging of gradients and eddy diffusivities, calculation of average fluxes

As was explained in the manuscript, the concentration gradients were averaged over 50 to 140 hrs and the half-hour eddy diffusivity values obtained from the EC measurements were averaged over the same periods for the purpose of flux calculation. In general, the micrometeorological measurements are used over half-hourly to hourly time period (let us say T) for calculation of turbulent fluxes. In order to obtain correct time averaged flux (over 50 to 140 hrs) one should ideally calculate fluxes over stationary time periods (1/2 to 1 hr) and then average the fluxes. Thus, in general $\left\langle F(z) \right\rangle = -\left\langle K_z \dfrac{\partial \overline{C}}{\partial z} \right\rangle \neq -\left\langle K_z \right\rangle \left\langle \dfrac{\partial \overline{C}}{\partial z} \right\rangle$ (here the angle brackets denote averaging over non-stationary, long period of time).

However, if the eddy diffusivity K_z is relatively constant, then the average fluxes obtained by both ways would be close. If K_z varies significantly during 50 to 140 hrs used for averaging of profiles, there is a risk of obtaining biased time-averaged flux estimates. As shown above, the eddy diffusivity is proportional to u* and under higher wind (also proportional to u*) the eddy diffusivity is expected to be higher and the concentration gradients smaller, respectively.

Below a simplified reasoning for evaluation of the average flux uncertainty follows. Let us assume ϕ_c is constant over time (e.g. near-neutral conditions), but u_* varies such that pdf of u_* corresponds to observations. Then assuming that the flux (source strength) is constant over time (F_0), it can be presented for certain friction velocity value u_{*0} by $F_0 = -\frac{k\ z\ u_{*0}}{\phi_c} \left.\frac{\partial \overline{C}}{\partial z}\right|_0$. For arbitrary $u_* = u_{*0}\ \varepsilon$ (with ε representing a dimensionless random variable) the flux reads as $F_0 = -\frac{k\ z\ u_{*0}\ \varepsilon}{\phi_c} \frac{1}{\varepsilon} \left.\frac{\partial \overline{C}}{\partial z}\right|_0$. The error introduced by averaging diffusivities $\langle K_z \rangle = \left\langle \frac{k\ z\ u_{*0}\ \varepsilon}{\phi_c} \right\rangle$ and gradients $\left\langle \frac{\partial \overline{C}}{\partial z} \right\rangle = \left\langle \frac{1}{\varepsilon} \left.\frac{\partial \overline{C}}{\partial z}\right|_0 \right\rangle$ separately over time, and then calculating the product as the flux estimate $F_{0E} = -\langle K_z \rangle \left\langle \frac{\partial \overline{C}}{\partial z} \right\rangle$, can be represented by $\frac{\Delta F_{0E}}{F_0} = \frac{F_{0E} - F_0}{F_0} = \langle \varepsilon \rangle \left\langle \frac{1}{\varepsilon} \right\rangle - 1$. Such error is expected to be in the order of ten to a few tens of percents, which can be considered perhaps as the underestimated uncertainty due to over-simplification of the matter (assuming constant source and near-neutral stability).

The authors should evaluate the error introduced by averaging of profiles and K_z over non-stationary period of time or convince by some reasoning (e.g. based on the expected source and K_z behaviour over the averaging period) that the error is expected to be reasonably small.