Reply to "Comment on 'Flow-distributed oscillations: Stationary chemical waves in a reacting flow' "

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Stationary waves (wavelength λ) are the necessary result of spatial recurrence of phase in the open flow (rate v) of an oscillating medium with fixed inflow boundary conditions. Any nonlinear dependence $\lambda(v)$ on flow is the result of dispersion that is implicit in $\lambda = v/\omega(v)$, where $\omega(v)$ is the oscillation frequency in a reference frame moving with the flow. The flow-distributed oscillator mechanism extends thus from the purely kinematic limit ω=const to the case where a nonlinear dependence $\lambda(v)$ is subsumed by the dispersion relationship $\omega(v)$.

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Andrésen *et al.* predicted [1] that stationary concentration waves may arise in a flow of an oscillating medium with constant inflow boundary forcing when all species have equal flow and diffusion coefficients. We suggested [2] that these waves arise by a mechanism that is essentially kinematic through the spatial recurrence of the phase of a flow-distributed oscillator (FDO). We observed a linear dependence of the wavelength $\lambda(v)$ on flow-rate v. This is described by the relation $\lambda = v/\omega$ if the oscillation frequency is independent of flow, as expected in the purely kinematic limit of negligible diffusion $D \to 0$. However, the FDO mechanism is also valid in the presence of diffusion $D \neq 0$, where the oscillation frequency $\omega(\lambda)$ becomes, through dispersion of the nonlinear waves, a function of wavelength $\lambda(v,D)$. As a result, the wavelength

$$\lambda = v/\omega(\lambda) \qquad (1)$$

may, depending on the dispersion-relation $\omega(\lambda)$, be a nonlinear function of flow. Hence the description remains *essentially* kinematic, and any nonlinear dependence of $\lambda(v)$ is included implicitly through the dispersion-relationship $\omega(\lambda)$, where $\lambda = \lambda(v,D)$ [3]. The FDO mechanism is therefore fully consistent with the nonlinear dependence of wavelength on the flow rate in Fig. 1 of the preceding Comment, which the authors however adduce as evidence of its purported failure. When $\omega(v,D)$ is measured experimentally, the FDO mechanism also accounts *quantitatively* (unpublished results) for the upstream and downstream traveling waves observed with periodic boundary forcing and it captures the kinematic origin of pulsating waves [4].

Consider a stationary-wave profile generated in a flow of an oscillating medium with constant boundary forcing as shown in Fig. 1. Since all species have identical transport coefficients, it is appropriate to consider a volume element (circles in Fig. 1) that is convected downstream at the flow rate v. In the comoving reference frame $x = vt$ its temporal evolution is given by

$$u_t = f(u) + Du_{xx} = g(u,D,v), \quad u(t=0,x=0) = u_0, \qquad (2)$$

where $f(u)$ describes the kinetics of $u = u_1, \ldots, u_n$. The constant initial condition arises from constant boundary forcing and the dependence on v is introduced through the term Du_{xx}, as explained below. While a volume element is convected downstream, it evolves from the initial condition u_0 to its asymptotic, stable solution, i.e., $u(t) \to U(t';D,v)$ for $t \to \infty$, if such a solution exists. If U describes a limit cycle with period $T = 1/\omega$, i.e., $U(t') = U(t'+T)$, then the volume element eventually oscillates with frequency ω and its phase recurs at equidistant points separated by v/ω. If all volume elements that subsequently enter the flow evolve following $u(t)$, the phase recurrence of the individual volume elements amounts to a stationary phase wave. Conversely, if a stationary space-periodic wave is established, there must exist a stable periodic solution $U(t';v,D)$ to Eq. (2) and the formation of a stationary phase wave is *necessarily* the result of this oscillation being distributed through space by the flow.

When the wave-profile $u(x)$ is stationary in the fixed reference frame (Fig. 1) it is generated by the temporal evolution of u in the comoving reference frame. The term Du_{xx} can thus be replaced by $D/v^2 u_{tt}$. In the kinematic limit $R \equiv D/v^2 \to 0$ of negligible diffusion, the solution of Eq. (2) is

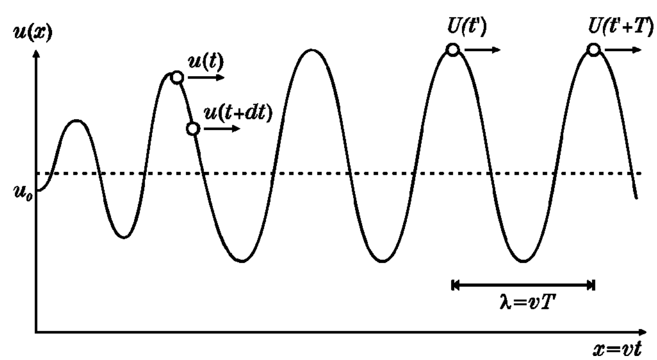

FIG. 1. Stationary concentration profile $u(x)$ (full line) generated by the temporal evolution of a volume element (circles) moving downstream with the flow rate v ($t = x/v$). Its evolution form the initial condition $u(0) = u_0$ is $u(t)$ that approaches a stable limit cycle $U(t') = U(t'+T)$ for $t \to \infty$. $U(t')$ is the flow-distributed oscillator that establishes a stationary space-periodic wave with wavelength $\lambda = vT$. Broken line represents a homogeneous stationary state that has become unstable through a Hopf bifurcation. The stable limit cycle created in this bifurcation coincides with $U(t')$ in the absence of diffusion.

the same as that of the well-mixed batch system $(D=0)$ that oscillates with a frequency ω' independently of the flow rate [2]. Diffusion is not required for the formation of stationary waves and plays a subordinate, but noticeable, role since it only alters the frequency of the FDO [2]. However, diffusion has to be sufficiently weak to remain in the supercritical $v>v_c$ FDO domain. In the subcritical $v<v_c$ domain, where stationary waves are no longer formed and the FDO mechanism no longer applies, the system should be regarded as a reaction-diffusion system perturbed by a flow and boundary forcing. Although the FDO mechanism does not capture the cessation of stationary waves and bifurcation to traveling waves at $v=v_c$, it adequately describes the essential physics of the supercritical wave phenomena.

While λ depends linearly on v when diffusion is negligible $(\omega=\omega'=\text{const})$, the dependence is generally nonlinear due to dispersion, as shown in Fig. 1 of the preceding Comment. The figure also shows that $\lambda(v)$ is linear when v is close to v_c, i.e., $\lambda(v)$ has a critical exponent of unity, and that this linearity persists over an extended range of v if diffusion is turbulent $D=cv$ $(R=c/v)$. This is what we find in our experiment that was done in a packed bed reactor, i.e., turbulent diffusion, close to v_c $(v_{max}\approx 2v_c)$. Thus, our experimental observations as well as the FDO mechanism are readily reconciled with the findings by Andrésen *et al.*, if one keeps in mind that $\omega(v,D)$ is the actual, experimentally observable frequency recorded in downstream moving volume elements, which accounts implicitly for dispersion.

[1] P. Andresén, M. Bache, E. Mosekilde, G. Dewel, and P. Borckmans, Phys. Rev. E **60**, 297 (1999).
[2] M. Kærn and M. Menzinger, Phys. Rev. E **60**, 3471 (1999).
[3] The frequency ω can either be obtained theoretically from the dispersion relation or measured experimentally.
[4] M. Kærn and M. Menzinger, Phys. Rev. E **61**, 3334 (2000).