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Spectral analysis and computation of effective diffusivities in space-time periodic incompressible flows

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The diffusive transport of passive tracers or particles can be enhanced by incompressible, turbulent flow fields. Analyzing the effective behavior is a challenging problem with theoretical and practical importance in many areas of science and engineering, ranging from the transport of mass, heat, and pollutants in geophysical flows to sea ice dynamics and turbulent combustion. The long time, large scale behavior of such systems is equivalent to an enhanced diffusion process with an effective diffusivity tensor D^*. Two different formulations of integral representations for D^* were developed for the case of *time-independent* fluid velocity fields, involving spectral measures of *bounded* self-adjoint operators acting on vector fields and scalar fields, respectively. Here, we extend both of these approaches to the case of *space-time periodic* velocity fields, allowing for chaotic dynamics, providing rigorous integral representations for D^* involving spectral measures of *unbounded* self-adjoint operators. We prove the different formulations are equivalent. Their correspondence follows from a one-to-one isometry between the underlying Hilbert spaces. We also develop a Fourier method for computing D^*, which captures the phenomenon of residual diffusion related to Lagrangian chaos of a model flow. This is reflected in the spectral measure by a concentration of mass near the spectral origin.

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1. Introduction

The long time, large scale motion of diffusing particles or tracers being advected by an incompressible flow field is equivalent to an enhanced diffusion process [99] with an effective diffusivity tensor D^*. Describing the associated transport properties is a challenging problem with a broad range of scientific and engineering applications, such as stellar convection [47, 86, 20, 21, 19], turbulent combustion [3, 15, 98, 105, 82, 107], and solute transport in porous media [12, 13, 104, 41, 48, 51, 49]. Time-dependent flows can have fluid velocity fields with chaotic dynamics, which gives rise to turbulence that greatly enhances the mixing, dispersion, and large scale transport of diffusing scalars. Here, we develop a mathematical framework that provides an analytic representation of D^* for such time-dependent, chaotic flows. This representation is given in terms of a Stieltjes integral involving the spectral measure of an *unbounded* self-adjoint operator and the molecular diffusion constant ε. We demonstrate that this approach provides an effective method for computing D^* for a model, chaotic flow.

1.1. Advection enhanced diffusion in the climate system

In the climate system [25, 40], turbulence plays a key role in transporting mass, heat, momentum, energy, and salt in geophysical flows [69]. Turbulence enhances the dispersion of atmospheric gases [27] such as ozone [43, 83, 84, 85] and pollutants [24, 11, 91], as well as atmosphere-ocean transfers of carbon dioxide and other climatically important trace gas fluxes [109, 8]. Longitudinal dispersion of passive scalars in oceanic flows can be enhanced by horizontal turbulence due to shearing of tidal currents, wind drift, or waves [108, 50, 17]. Chaotic motion of time-dependent fluid velocity fields causes instabilities in large scale ocean currents, generating geostrophic eddies [31] which dominate the kinetic energy of the ocean [32]. Geostrophic eddies greatly enhance [31] the meridional mixing of heat, carbon and other climatically important tracers, typically more than one order of magnitude greater than the mean flow of the ocean [94]. Eddies also impact heat and salt budgets through lateral fluxes and can extend the area of high biological productivity offshore by both eddy chlorophyll advection and eddy nutrient pumping [22].

In sea ice dynamics, where the ice cover couples the atmosphere to the polar oceans [102], the transport of sea ice can also be enhanced by eddie fluxes and large scale coherent structures in the ocean [103, 54]. In sea ice thermodynamics, the temperature field of the atmosphere is coupled to the temperature field of the ocean through sea ice, which is a composite of pure ice with brine inclusions whose volume fraction and connectedness depend strongly on temperature [100, 37, 36]. Convective brine flow through the porous microstructure can enhance thermal transport through the sea ice layer [55, 106, 52].

Both numerical and observational studies of scalar transport have suggested that tracers are advected over large scales by a fluid velocity field that is different from the mean flow [79]. This suggests that the effective diffusivity tensor D^* should be spatially and possibly also temporally inhomogeneous [79]. The mixing of eddy fluxes is typically non-divergent and unable to affect the evolution of the mean flow [66], and do not alter the tracer moments [39]. In this sense, the mixing is non-dissipative, reversible, and sometimes referred to as stirring [26, 39]. It has been noted in various geophysical contexts [84, 85] that eddy-induced skew-diffusive tracer fluxes directed normal to the tracer gradient [66] are generally equivalent to *antisymmetric* components in the effective diffusivity tensor D^*, while the *symmetric* part of D^* represents irreversible diffusive effects [87, 92, 39] directed down the tracer gradient. Motivated by these observations, in the ensuing sections we provide analytic representations for both the *symmetric* and *antisymmetric* components of D^*.

1.2. Mathematical characterization of effective diffusivity

Due to the computational intensity of detailed climate models [40, 102, 73], a coarse resolution is necessary in numerical simulations and *parameterization* is used to help resolve sub-grid scale processes, such as turbulent entrainment-mixing processes in clouds [53], atmospheric boundary layer turbulence [18], atmosphere-surface exchange over the sea [28] and sea ice [93, 1, 2, 101], and eddies in the ocean [60, 35]. In this way, only the effective or averaged behavior of these sub-grid processes are included in the models. Here, we study the effective behavior of advection enhanced diffusion by time-dependent fluid velocity fields, with possibly chaotic dynamics, which gives rise to such a parameterization, namely, the effective diffusivity tensor D^* of the flow.

In recent decades, a broad range of mathematical techniques have been developed which reduce the analysis of enhanced diffusive transport by complex fluid velocity fields with rapidly varying structures in both space and time, to solving averaged or *homogenized* equations that do not have rapidly varying data, and involve an effective parameter [75, 61, 9, 14, 29, 74, 30, 58, 79, 80, 23, 42, 44, 56, 57, 107]. Motivated by [76], it was shown in [61] that the homogenized behavior of the advection-diffusion equation with a random, time-independent, incompressible, mean-zero fluid velocity field, is given by an inhomogeneous diffusion equation involving the symmetric part of an effective diffusivity tensor D^*. Moreover, a rigorous representation of D^* was given in terms of an auxiliary *cell or corrector problem* involving a curl-free random field [61]. We stress that the effective diffusivity tensor D^* is not symmetric in general. However, only its symmetric part appears in the homogenized equation for *this* formulation of the effective transport properties of advection enhanced diffusion [61].

The incompressibility condition of the time-independent fluid velocity field was used in [4, 5] to transform the cell problem in [61] into the quasi-static limit of Maxwell's equations [46, 38], which describe the transport properties of an electromagnetic wave in a composite material [68]. The analytic continuation method for representing transport in composites [38] provides Stieltjes integral representations for the bulk transport coefficients of composite media, such as electrical conductivity and permittivity, magnetic permeability, and thermal conductivity [68]. This method is based on the spectral theorem [97, 88] and a resolvent formula for, say, the electric field, involving a random self-adjoint operator [38, 72] or matrix [70]. Based on the analytic continuation method [38], in [4, 5] the cell problem for the advection diffusion equation was transformed into a resolvent formula involving a *bounded* self-adjoint operator, acting on the Hilbert space of curl-free random vector fields. This, in turn, led to a Stieltjes integral representation for the symmetric part of the effective diffusivity tensor D^*, involving the Péclet number Pe of the flow and a *spectral measure* $\boldsymbol{\mu}$ of the operator [4, 5]. A key feature of the method is that parameter information in Pe is *separated* from the complicated geometry of the time-independent flow, which is encoded in the measure $\boldsymbol{\mu}$. This property led to rigorous bounds [5] for the diagonal components of D^*. Bounds for D^* can also be obtained using variational methods [5, 29, 74, 30].

The mathematical framework developed in [61] was also adapted [79, 56] to the case of a periodic, time-dependent, incompressible fluid velocity field with *non-zero* mean. The velocity field was modeled as a superposition of a large-scale mean flow with small-scale periodically oscillating fluctuations. It was shown [79] that, depending on the strength of the fluctuations relative to the mean flow, the effective diffusivity tensor D^* can be constant or a function of both space and time. When D^* is constant, only its symmetric part appears in the homogenized equation as an enhancement in the diffusivity. However, when D^* is a function of space and time, its antisymmetric part also plays a key role in the homogenized equation. In particular, the symmetric part of D^* appears as an enhancement in the diffusivity, while both the symmetric and antisymmetric parts of D^* contribute to an effective drift in the homogenized equation. The effective drift due to the antisymmetric part is purely sinusoidal, thus divergence-free [79]. This is consistent with what has been observed in geophysical flows in the climate system, as discussed in the final paragraph of Section 1.1.

In an alternate formulation of the effective parameter problem based on [12], the cell problem discussed in [79] was transformed into a resolvent formula involving a self-adjoint operator acting on a Sobolev space [63, 33] of spatially periodic scalar fields, which is also a Hilbert space. In the case where the mean flow and periodic fluctuations are time-independent, the self-adjoint operator is compact [12], hence *bounded* [95]. This led to a discrete Stieltjes integral representation for the antisymmetric part of D^*, involving the Péclet number of the steady flow and a spectral measure of the operator.

The incompressibility of the fluid velocity field is a central property of the mathematical frameworks described above. However, these results were extended in [62] to weakly compressible, anelastic, stratified, time-independent, fluid velocity fields. Homogenization of the convection-reaction-diffusion equation with a compressible velocity field is treated in [77].

1.3. Summary of results

Here, we generalize both of the approaches described in [4, 5] and [79] to the case of an incompressible, periodic, *time-dependent* fluid velocity field, allowing for chaotic dynamics. In particular, for each approach, we provide Stieltjes integral representations for both the symmetric and antisymmetric parts of the effective diffusivity tensor D^*, involving a spectral measure of a self-adjoint operator. In this time-dependent setting, the underlying operator becomes *unbounded*. The spectral theory of unbounded operators is more subtle and technically challenging than the spectral theory of bounded operators, since the domain of an unbounded operator and its adjoint plays a central role in the spectral characterization of the operator. Neglecting such important mathematical details, the Stieltjes integral representation for D^* given in [4, 5] was extended to the time-dependent setting in [6]. Here, we provide mathematically rigorous formulations of Stieltjes integral representations for D^* in the time-dependent, unbounded operator setting. Moreover, we prove that the two approaches in [4, 5] and [79] are equivalent in this setting, and that their correspondence follows from a one-to-one isometry between the underlying Hilbert spaces. We also establish a direct correspondence between the effective parameter problem for D^* and the analogous effective parameter problem arising in the analytic continuation method for composite materials.

In over 25 years since the first derivation [4] of an integral representation for the effective diffusivity tensor D^*, analytical calculations of the underlying spectral measure have been obtained only for a handful of simple flows, such as shear flow [5], and numerical computations of the effective behavior based on this powerful representation have apparently not been attempted. To help overcome this limitation, we develop a Fourier method for the computation of D^*. In particular, we compute the effective properties for the following space-time periodic flow in two spatial dimensions, with $\boldsymbol{x} = (x, y)$,

$$\boldsymbol{u}(t, \boldsymbol{x}) = (\cos y, \cos x) + \theta \, \cos t \, (\sin y, \sin x), \quad \theta \in (0, 1]. \tag{1}$$

The steady part $(\cos y, \cos x)$ of the flow is subject to a time-periodic perturbation that gives rise to a transition to Lagrangian chaos for $\theta > 0$ [14, 110]. In a study of *residual diffusivity* [14, 110] for the advection dominated regime, we shall compare our computations of the effective diffusivity for the steady $\theta = 0$ and dynamic $\theta = 1$ settings.

The rest of the paper is organized as follows. In Section 2, the theory of homogenization for the advection-diffusion equation for space-time periodic flows is reviewed. Novel Stieltjes integral representations for the effective diffusivity tensor D^* are also obtained for a large class of space-time periodic fluid velocity fields, involving a spectral measure of an *unbounded* self-adjoint operator. In Section 3, we provide a rigorous mathematical framework for the computation of the discrete part of the spectral measure μ and integral representation for D^*, providing a rigorous lower bound for D^*. In particular, we use Fourier methods to transform the eigenvalue problem for the self-adjoint operator involving the space-time periodic fluid velocity field in equation (1) into an infinite system of algebraic equations. This framework is employed in Section 4 to compute the discrete component of D^* for the velocity field in (1), for both the time-independent $\theta = 0$ and time-dependent $\theta = 1$ settings.

Our computations highlight that the behavior of the measure near the spectral origin governs the behavior of the effective diffusivity in the advection dominated regime of small molecular diffusion. In particular, we demonstrate that for $\theta = 0$ there is a *spectral gap* in the measure near a limit point at the spectral origin, giving rise to the known vanishing asymptotic behavior of 2D cell flows [29, 74]. However in the time dependent setting, a strong concentration of measure mass near the spectral origin gives rise to the phenomenon of residual diffusivity in the limit of vanishing molecular diffusion.

Technical background information and proofs of the key results of the paper are deferred to the appendices. The spectral theory of unbounded self-adjoint operators in Hilbert space is reviewed in Appendix A and Appendix B. Two mathematical formulations of the effective parameter problem for advection enhanced diffusion are presented in Appendix C.1 and Appendix C.2, leading to novel integral representations for the symmetric and antisymmetric components of the effective diffusivity tensor. In Appendix D we use powerful methods of functional analysis to prove that the two approaches are equivalent, which follows from a one-to-one isometry between the associated Hilbert spaces. In Appendix E we derive an explicit formula for the discrete component of the spectral measure, which is employed in our numerical computations.

2. Effective transport by advective-diffusion

The density ϕ of a cloud of passive tracer particles diffusing along with molecular diffusivity ε and being advected by an incompressible velocity field $\boldsymbol{u}$ satisfies the advection-diffusion equation

$$\partial_t\phi(t,\boldsymbol{x}) = \boldsymbol{u}(t,\boldsymbol{x})\cdot\boldsymbol{\nabla}\phi(t,\boldsymbol{x}) + \varepsilon\Delta\phi(t,\boldsymbol{x}), \quad \phi(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}), \tag{2}$$

for $t > 0$ and $\boldsymbol{x} \in \mathbb{R}^d$. Here, the initial density $\phi_0(\boldsymbol{x})$ and the fluid velocity field $\boldsymbol{u}$ are assumed given, and $\boldsymbol{u}$ satisfies $\boldsymbol{\nabla}\cdot\boldsymbol{u} = 0$. In equation (2), the molecular diffusion constant $\varepsilon > 0$, d is the spatial dimension of the system, ∂_t denotes partial differentiation with respect to time t, and $\Delta = \boldsymbol{\nabla}\cdot\boldsymbol{\nabla} = \nabla^2$ is the Laplacian. Moreover, $\boldsymbol{\psi}\cdot\boldsymbol{\varphi} = \boldsymbol{\psi}^T\overline{\boldsymbol{\varphi}}$, $\boldsymbol{\psi}^T$ denotes transposition of the vector $\boldsymbol{\psi}$, and $\overline{\boldsymbol{\varphi}}$ denotes component-wise complex conjugation, with $\boldsymbol{\psi}\cdot\boldsymbol{\psi} = |\boldsymbol{\psi}|^2$. Later, we will extensively use this form of the dot product over complex fields, with built in complex conjugation. However, we stress that all quantities considered in this section are *real-valued*.

We consider enhanced diffusive transport by a periodic fluid velocity field and non-dimensionalize equation (2) as follows. Let L and T be typical length and time scales associated with the problem of interest. Mapping to the non-dimensional variables $t \mapsto t/T$ and $\boldsymbol{x} \mapsto \boldsymbol{x}/L$, one finds that ϕ satisfies the advection-diffusion equation in (2) with a non-dimensional molecular diffusivity $\varepsilon \mapsto T\,\varepsilon/L^2$ and velocity field $\boldsymbol{u} \mapsto T\,\boldsymbol{u}/L$. There are several different non-dimensionalizations possible for the advection-diffusion equation. A detailed discussion of various non-dimensionalizations involving the Strouhal number, the Péclet number, and the periodic Péclet number is given in [62, 56]. Here, we focus on the long time, large scale transport characteristics of equation (2) as a function of ε. To this end, for the case of a time-dependent flow we simply take T to be the temporal periodicity of the velocity field $\boldsymbol{u}$ and assume that the spatial periodicity of $\boldsymbol{u}$ is L in all spatial dimensions, i.e.,

$$(3) \qquad \boldsymbol{u}(t+T,\boldsymbol{x}) = \boldsymbol{u}(t,\boldsymbol{x}), \qquad \boldsymbol{u}(t,\boldsymbol{x}+L\,\boldsymbol{e}_j) = \boldsymbol{u}(t,\boldsymbol{x}), \quad j=1,\dots,d,$$

where $\boldsymbol{e}_j$ is a standard basis vector in the jth direction. In the case of a time-independent spatially periodic flow, a natural choice for L and T is, respectively, the maximum cell period and $T = L/\langle|\boldsymbol{u}|^2\rangle^{1/2}$, yielding the non-dimensional quantities $\varepsilon \mapsto \varepsilon/(L\,\langle|\boldsymbol{u}|^2\rangle^{1/2})$ and $\boldsymbol{u} \mapsto \boldsymbol{u}/\langle|\boldsymbol{u}|^2\rangle^{1/2}$.

2.1. Mean-zero flow

In this section we will discuss the effective transport properties of advection enhanced diffusion, as described by the advection diffusion equation in (2). We will assume in this section that the fluid velocity field is mean-zero. The effects of a large-scale mean flow will be discussed in Section 2.2.

The long time, large scale dispersion of diffusing tracer particles being advected by an incompressible fluid velocity field is equivalent to an enhanced diffusion process [99] with an effective diffusivity tensor D^*. In recent decades, methods of homogenization theory [61, 29, 74, 56] have been used to provide an explicit representation for D^*. In particular, these methods have demonstrated that the averaged or *homogenized* behavior of the advection-diffusion equation in (2), with space-time periodic velocity field $\boldsymbol{u}$, is determined by a diffusion equation involving an averaged scalar density $\bar{\phi}$ and an effective diffusivity tensor D^* [56]

$$(4) \qquad \partial_t\bar{\phi}(t,\boldsymbol{x}) = \boldsymbol{\nabla}\cdot[\mathsf{D}^*\boldsymbol{\nabla}\bar{\phi}(t,\boldsymbol{x})], \quad \bar{\phi}(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}).$$

Equation (4) follows from the assumption that the initial tracer density ϕ_0 varies slowly relative to the variations of the fluid velocity field $\boldsymbol{u}$ [61, 30, 56]. This information is incorporated into equation (2) by introducing a small dimensionless parameter $\delta \ll 1$ and writing [61, 30, 56]

$$(5) \qquad \phi(0,\boldsymbol{x}) = \phi_0(\delta\boldsymbol{x}).$$

Anticipating that ϕ will have diffusive dynamics as $t \to \infty$, space and time are rescaled according to the standard diffusive relation

$$\boldsymbol{\xi} = \boldsymbol{x}/\delta, \quad \tau = t/\delta^{\gamma}, \qquad \gamma = 2. \tag{6}$$

The rescaled form of equation (2) is given by [56]

$$\partial_t \phi^{\delta}(t,\boldsymbol{x}) = \delta^{-1}\boldsymbol{u}(t/\delta^2, \boldsymbol{x}/\delta)\boldsymbol{\cdot}\boldsymbol{\nabla}\phi^{\delta}(t,\boldsymbol{x}) + \varepsilon\Delta\phi^{\delta}(t,\boldsymbol{x}), \quad \phi^{\delta}(0,\boldsymbol{x}) = \phi_0(\boldsymbol{x}), \tag{7}$$

where we have denoted $\phi^{\delta}(t,\boldsymbol{x}) = \phi(t/\delta^2, \boldsymbol{x}/\delta)$. The convergence of ϕ^{δ} to $\bar{\phi}$ can be rigorously established in the following sense [56]

$$\lim_{\delta\to 0} \sup_{0\le t\le t_0} \sup_{\boldsymbol{x}\in\mathbb{R}^d} |\phi^{\delta}(t,\boldsymbol{x}) - \bar{\phi}(t,\boldsymbol{x})| = 0, \tag{8}$$

for every finite $t_0 > 0$, provided that ϕ_0 and $\boldsymbol{u}$ obey some mild smoothness and boundedness conditions, and that $\boldsymbol{u}$ is *mean-zero* (also see [81]). We will discuss the consequences of a fluid velocity field $\boldsymbol{u}$ with a large scale mean flow in Section 2.2.

An explicit representation of the effective diffusivity tensor D^* is given in terms of the (unique) mean zero, space-time periodic solution χ_j of the following *cell problem* [14, 56],

$$\partial_\tau \chi_j(\tau,\boldsymbol{\xi}) - \varepsilon\Delta_{\xi}\chi_j(\tau,\boldsymbol{\xi}) - \boldsymbol{u}(\tau,\boldsymbol{\xi})\boldsymbol{\cdot}\boldsymbol{\nabla}_{\xi}\chi_j(\tau,\boldsymbol{\xi}) = u_j(\tau,\boldsymbol{\xi}), \tag{9}$$

where the subscript ξ in Δ_ξ and $\boldsymbol{\nabla}_\xi$ indicates that differentiation is with respect to the fast variable $\boldsymbol{\xi}$ defined in equation (6). The components D^*_{jk}, $j,k = 1,\ldots,d$, of the matrix D^* are given by [61, 29, 74, 56]

$$\mathsf{D}^*_{jk} = \varepsilon\delta_{jk} + \langle u_j \chi_k\rangle, \tag{10}$$

where δ_{jk} is the Kronecker delta and u_j is the jth component of the vector $\boldsymbol{u}$. The averaging $\langle\cdot\rangle$ in (10) is with respect to the fast variables defined in equation (6). The averaging is over the bounded sets $\mathcal{T} \subset \mathbb{R}$ and $\mathcal{V} \subset \mathbb{R}^d$, with $\tau \in \mathcal{T}$ and $\boldsymbol{\xi} \in \mathcal{V}$, which define the space-time period cell ($(d+1)$–torus) $\mathcal{T}\times\mathcal{V}$. For example, in Section 4 we compute D^* for the fluid velocity field $\boldsymbol{u}$ in (1) with temporal periodicity $\mathcal{T} = [0, 2\pi]$ and spatial periodicity $\mathcal{V} = [0, 2\pi]^d$, with $d = 2$. In the case of a time-dependent fluid velocity field, $\langle\cdot\rangle$ denotes space-time averaging over $\mathcal{T}\times\mathcal{V}$. In the special case of a time-independent fluid velocity field, the function χ_j is time-independent and satisfies equation (9) with $\partial_\tau\chi_j \equiv 0$, and $\langle\cdot\rangle$ in (10) denotes spatial averaging over $\mathcal{V}$ [29, 74, 56].

2.2. The effect of large scale mean flow

The periodic homogenization theorem summarized by equations (3)–(10) depends on the detailed nature of the fluid velocity field $\boldsymbol{u}$. It also depends on the temporal scaling used [12, 79, 56], i.e., what value of γ is used in equation (6). However, the mathematical structure of the cell problem in (9) and the functional form of D^* shown in equation (10) remain unchanged for the space-time periodic setting. In order to illustrate the rich behaviors that can arise in the effective diffusivity tensor D^* for more general velocity fields and alternate temporal scalings, we now discuss some key variations of the theory described above.

In general, the effective diffusivity tensor D^* has a symmetric S^* and antisymmetric A^* part defined by

$$(11)\qquad \mathsf{D}^* = \mathsf{S}^* + \mathsf{A}^*, \qquad \mathsf{S}^* = \frac{1}{2}\left(\mathsf{D}^* + [\mathsf{D}^*]^T\right), \quad \mathsf{A}^* = \frac{1}{2}\left(\mathsf{D}^* - [\mathsf{D}^*]^T\right),$$

where $[\mathsf{D}^*]^T$ denotes transposition of the matrix D^*. Denote by S^*_{jk} and A^*_{jk}, $j, k = 1, \ldots, d$, the components of S^* and A^* in (11). When the fluid velocity field is mean-zero and divergence-free, as discussed above, then equation (8) holds and the effective diffusivity tensor D^* defined in (10) is constant [56]. Consequently, only the symmetric part of D^* plays a role in the effective transport equation shown in (4) [79].

Now consider the more general, divergence-free fluid velocity field

$$(12)\qquad \boldsymbol{u}(t, \boldsymbol{x}) = \delta \boldsymbol{u}_0(\delta^2 t, \delta \boldsymbol{x}) + \boldsymbol{u}_1(t, \boldsymbol{x}),$$

which is the superposition of a *weak*, large-scale mean flow $\delta \boldsymbol{u}_0(\delta^2 t, \delta \boldsymbol{x})$ that varies on large spatial and slow time scales, with a mean-zero periodic flow $\boldsymbol{u}_1(t, \boldsymbol{x})$ that rapidly fluctuates in space and time [56]. If $\boldsymbol{u}_0(t, \boldsymbol{x})$ is smooth and bounded, the homogenization theorem for purely periodic velocity fields discussed above can be rigorously extended to the present setting and the effective transport equation in (4) is replaced by [56]

$$(13)\qquad \partial_t \bar{\phi}(t, \boldsymbol{x}) = \boldsymbol{u}_0(t, \boldsymbol{x}) \cdot \boldsymbol{\nabla} \bar{\phi}(t, \boldsymbol{x}) + \boldsymbol{\nabla} \cdot [\mathsf{D}^* \boldsymbol{\nabla} \bar{\phi}(t, \boldsymbol{x})], \quad \bar{\phi}(0, \boldsymbol{x}) = \phi_0(\boldsymbol{x}),$$

which includes an advective enhancement in transport by the large-scale mean flow $\boldsymbol{u}_0$ [56]. In this case, the effective diffusivity tensor D^* is completely independent of the mean flow $\boldsymbol{u}_0$, and is determined by the same formula in equation (10) and the same cell problem in (9) with $\boldsymbol{u}$ replaced by the *mean-zero* velocity field $\boldsymbol{u}_1$ [56]. Consequently, D^* is again constant and only the symmetric part of D^* plays a role in the effective transport equation shown in (13).

In [79], D^* was studied for the divergence-free fluid velocity field,

$$\boldsymbol{u}(t,\boldsymbol{x}) = \boldsymbol{u}_0(t,\boldsymbol{x}) + \delta^\alpha \boldsymbol{u}_1(t/\delta^\gamma, \boldsymbol{x}/\delta), \tag{14}$$

for a broad range of scaling parameters γ and α. The parameter γ controls the separation of time scales while α determines the strength of the small scale periodic fluctuations $\boldsymbol{u}_1$ relative to the mean flow $\boldsymbol{u}_0$. There are three distinct behaviors that arise as the values of α and γ vary, and the function χ_j satisfying an analogue of the cell problem in (9) can be time-dependent or time-independent ($\partial_\tau \chi_j \equiv 0$) [79]. However, regardless of the values of α and γ studied in [79], when the mean flow is weak compared to the fluctuations, to leading order, D^* is constant and independent of the mean flow, which only determines the transport velocity on large length and long time scales, similar to equation (13). Consequently, only the symmetric part of D^* plays a role in the effective transport equation, which is similar to the effective transport equation in (13) [79]. However we emphasize that in all three cases, the components D^*_{jk} of the effective diffusivity tensor are given by a formula analogous to equation (10) and the structure of the cell problem is analogous to equation (9), where the velocity field component arising the right side of the cell problem is *mean-zero*.

The effective diffusivity tensor D^* being constant is *not* consistent with measurements and numerical simulations of passive tracer transport in the ocean and the atmosphere, as we discussed in the final paragraph of Section 1.1. However, when the fluid velocity field is active on both the slow and fast time scales, $\boldsymbol{u} = \boldsymbol{u}(t,\boldsymbol{x},t/\delta,\boldsymbol{x}/\delta)$, and the mean flow $\boldsymbol{u}_0(t,\boldsymbol{x}) = \langle \boldsymbol{u}(t,\boldsymbol{x},t/\delta,\boldsymbol{x}/\delta)\rangle$ is equal in strength or stronger than the periodic fluctuations, then the effective transport equation is analogous to equation (13) and D^* *is a function of both space and time* [79], $\mathsf{D}^* = \mathsf{D}^*(t,\boldsymbol{x})$. Consequently, in the effective transport equation, the *antisymmetric* part of $\mathsf{D}^*(t,\boldsymbol{x})$ contributes to a purely rotational (divergence-free) enhancement in advective transport, while the symmetric part of $\mathsf{D}^*(t,\boldsymbol{x})$ contributes to an enhancement in advective and diffusive transport [79]. This is consistent with observations and numerical simulations of geophysical flows in the climate system.

We emphasize that, in this formulation [79], the components $\mathsf{D}^*_{jk}(t,\boldsymbol{x})$, $j,k = 1,\dots,d$, of the effective diffusivity tensor are given by a formula that is analogous to equation (10). However, the function u_j appearing in (10) is replaced by the jth component of $\boldsymbol{u}(t,\boldsymbol{x},t/\delta,\boldsymbol{x}/\delta) - \boldsymbol{u}_0(t,\boldsymbol{x})$ which is *mean-zero*. Moreover, in this formulation [79], the cell problem is given by a formula that is analogous to equation (9). However the function u_j appearing on the right side of (9) is again replaced by the jth component of $\boldsymbol{u} - \boldsymbol{u}_0$ which is *mean-zero*. We show in Appendix C.1.2 that the essential conditions for Stieltjes integral representations for the symmetric S^* and antisymmetric A^* parts of D^* are: 1) the fluid velocity field $\boldsymbol{u}$ is *divergence free* and 2) the function u_j appearing in (10) and on the right side of equation (9) is *mean-zero*. Consequently, the Stieltjes integral representations for S^* and A^* discussed in the following section hold for all of the fluid velocity fields discussed in this section.

2.3. Integral representations for the effective diffusivity

In Appendix C.1 we provide a mathematically rigorous framework that leads to Stieltjes integral representations for the effective diffusivity tensor D^* for space-time periodic flows. This formulation is based on the spectral theorem for *unbounded* self-adjoint operators in Hilbert space. In Appendices A and B, we review the spectral theory of unbounded operators. In Appendix C we give two natural Hilbert space formulations of the effective parameter problem for D^* which lead to its integral representations. In Appendix D we prove that the two different formulations are equivalent.

In this section we summarize the results of Appendix C.1, which provide Stieltjes integral representations for both the symmetric S^* and antisymmetric A^* parts of D^*. Since the analysis in this section involves only the fast variables $(\tau, \boldsymbol{\xi})$ defined in equation (6), for notational simplicity, we will drop the subscripts ξ shown in equation (9) and use ∂_t to denote ∂_τ.

In Appendix C.1 we inserted the expression for u_j on the right side of (9) into equation (10), which leads to the following functional representations for the components S^*_{jk} and A^*_{jk}, $j, k = 1, \dots, d$, of S^* and A^* [79]

(15)
$$\mathsf{S}^*_{jk} = \varepsilon(\delta_{jk} + \langle \chi_j, \chi_k \rangle_{1,2}), \quad \mathsf{A}^*_{jk} = \langle A\chi_j, \chi_k \rangle_{1,2}\,, \quad A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla}).$$

Here, $\langle f, h \rangle_{1,2} = \langle \boldsymbol{\nabla} f \cdot \boldsymbol{\nabla} h \rangle$ is a Sobolev-type *sesquilinear* inner-product [63] and the operator $(-\Delta)^{-1}$ is based on convolution with respect to the Green's function for the Laplacian Δ [95]. Since the function χ_j is *real-valued* we have $\langle \chi_j, \chi_k \rangle_{1,2} = \langle \chi_k, \chi_j \rangle_{1,2}$, which implies that S^* is a symmetric matrix. The function $A\chi_j$ is also real-valued. We establish in Appendix C.1 that the operator A is skew-adjoint on a suitable Hilbert space, which implies that $\mathsf{A}^*_{kj} = \langle A\chi_k, \chi_j \rangle_{1,2} = -\langle \chi_k, A\chi_j \rangle_{1,2} = -\langle A\chi_j, \chi_k \rangle_{1,2} = -\mathsf{A}^*_{jk}$ which, in turn, implies that A^* is an antisymmetric matrix, hence $\mathsf{A}^*_{kk} = \langle A\chi_k, \chi_k \rangle_{1,2} = 0$.

Applying the linear operator $(-\Delta)^{-1}$ to both sides of the cell problem in equation (9) yields the following resolvent formula for χ_j

$$\chi_j = (\varepsilon + A)^{-1} g_j, \qquad g_j = (-\Delta)^{-1} u_j. \tag{16}$$

From equations (15) and (16) we have the following functional formulas for S^*_{jk} and A^*_{jk} involving the skew-adjoint operator A

$$\begin{aligned} \mathsf{S}^*_{jk} &= \varepsilon \left(\delta_{jk} + \langle (\varepsilon + A)^{-1} g_j, (\varepsilon + A)^{-1} g_k \rangle_{1,2} \right), \\ \mathsf{A}^*_{jk} &= \langle A(\varepsilon + A)^{-1} g_j, (\varepsilon + A)^{-1} g_k \rangle_{1,2}. \end{aligned} \tag{17}$$

Since A is a skew-adjoint operator, it can be written as $A = \imath M$ where M is a symmetric operator [97]. We demonstrate in Appendix C.1 that M is *self-adjoint* on an appropriate, dense subset of a Hilbert space.

The spectral theorem for self-adjoint operators states that there is a one-to-one correspondence between the self-adjoint operator M and a family of self-adjoint projection operators $\{Q(\lambda)\}_{\lambda\in\Sigma}$ — the resolution of the identity — that satisfies $\lim_{\lambda\to\inf\Sigma} Q(\lambda) = 0$ and $\lim_{\lambda\to\sup\Sigma} Q(\lambda) = I$ [97]. Here, Σ is the *spectrum* of the operator M, while 0 and I denote the null and identity operators. Define the *complex valued* function $\mu_{jk}(\lambda) = \langle Q(\lambda) g_j, g_k \rangle_{1,2}$, $j, k = 1, \ldots, d$, where $g_j = (-\Delta)^{-1} u_j$ is defined in (16). The real, $\operatorname{Re} \mu_{jk}(\lambda)$, and imaginary, $\operatorname{Im} \mu_{jk}(\lambda)$, parts of the function $\mu_{jk}(\lambda)$ are of bounded variation, and therefore have Stieltjes measures $\operatorname{Re} \mu_{jk}$ and $\operatorname{Im} \mu_{jk}$ associated with them [97]. The function $\mu_{kk}(\lambda)$ is positive and μ_{kk} is a positive measure, while $\operatorname{Re} \mu_{jk}$ and $\operatorname{Im} \mu_{jk}$, $j \neq k$, are signed measures. Given certain regularity conditions on the components u_j of the fluid velocity field $\boldsymbol{u}$, the functional formulas for S^*_{jk} and A^*_{jk} in (17) have the following Radon–Stieltjes integral representations, for all $0 < \varepsilon < \infty$ (see Appendix C.1 for details)

$$\mathsf{S}^*_{jk} = \varepsilon \left(\delta_{jk} + \int_{-\infty}^{\infty} \frac{\mathrm{dRe}\ \mu_{jk}(\lambda)}{\varepsilon^2 + \lambda^2} \right), \qquad \mathsf{A}^*_{jk} = -\int_{-\infty}^{\infty} \frac{\lambda\, \mathrm{dIm}\ \mu_{jk}(\lambda)}{\varepsilon^2 + \lambda^2}. \tag{18}$$

The integral formulas in (18) involve a spectral measure μ_{jk}, $j, k = 1, \ldots, d$, which has discrete and continuous components [88, 97]. The self-adjoint operator $M = -\imath A$ has *real* eigenvalues λ_l and orthonormal eigenfunctions φ_l, $l = 0, 1, 2, \ldots$, satisfying $M\varphi_l = \lambda_l \varphi_l$ and $\langle \varphi_i, \varphi_l \rangle_{1,2} = \delta_{il}$. In Appendix E.1 we employ an abstract mathematical framework to show the discrete parts $\tilde{\mathsf{S}}^*_{jk}$ and $\tilde{\mathsf{A}}^*_{jk}$ of the integral representations in (18) have the following series representations involving the λ_l and φ_l (see equation (A-66))

$$(19)\qquad \tilde{\mathsf{S}}^*_{jk} = \varepsilon\left(\delta_{jk} + \sum_{l=0}^{\infty}\frac{\operatorname{Re} m_{jk}(l)}{\varepsilon^2+\lambda_l^2}\right), \qquad \tilde{\mathsf{A}}^*_{jk} = -\sum_{l=0}^{\infty}\frac{\lambda_l \operatorname{Im} m_{jk}(l)}{\varepsilon^2+\lambda_l^2}\,.$$

Here, the *spectral weights* $m_{jk}(l)$ are given by (see equation (A-67))

$$(20)\qquad m_{jk}(l) = \langle g_j, \varphi_l\rangle_{1,2}\,\overline{\langle g_k, \varphi_l\rangle}_{1,2}, \qquad \langle g_j, \varphi_l\rangle_{1,2} = \langle u_j, \varphi_l\rangle = \langle u_j\,\overline{\varphi_l}\rangle.$$

In the setting of a time-independent fluid velocity field $\boldsymbol{u}=\boldsymbol{u}(\boldsymbol{x})$, the self-adjoint operator M is given by $M=-\imath(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]$. If $\boldsymbol{u}$ is smooth and uniformly bounded on $\mathcal{V}$, then M is a compact operator [12] and therefore has only discrete spectrum with a limit point at $\lambda=0$ [95, 88]. Consequently, the spectral measure μ_{jk} is purely discrete, hence $\mathsf{D}^*_{jk}\equiv\tilde{\mathsf{D}}^*_{jk}$. Since μ_{kk} is a *positive measure*, the discrete integral representation of $\tilde{\mathsf{S}}^*_{kk}$ in (19) provides a rigorous lower bound for the integral representation of S^*_{kk} in equation (18),

$$(21)\qquad \mathsf{S}^*_{kk} \geq \tilde{\mathsf{S}}^*_{kk}.$$

It is worth noting that using $\overline{\langle g_k,\varphi_l\rangle}_{1,2}=\langle\varphi_l, g_k\rangle_{1,2}$ and Dirac notation $\langle\boldsymbol{\nabla} g_j\cdot\boldsymbol{\nabla}\varphi_l\rangle=\langle\boldsymbol{\nabla} g_j|\boldsymbol{\nabla}\varphi_l\rangle$, we may formally write the spectral weights in equation (20) as

$$(22)\qquad \begin{aligned} m_{jk}(l) &= \langle\boldsymbol{\nabla} g_j|\boldsymbol{\nabla}\varphi_l\rangle\,\langle\boldsymbol{\nabla}\varphi_l|\boldsymbol{\nabla} g_k\rangle \\ &= \langle\boldsymbol{\nabla} g_j|\,\boldsymbol{\nabla}\,[\,|\varphi_l\rangle\,\langle\boldsymbol{\nabla}\varphi_l|\,\boldsymbol{\nabla}\,]\,|g_k\rangle \\ &= \langle\boldsymbol{\nabla} g_j|\boldsymbol{\nabla} Q_l\,g_k\rangle, \end{aligned}$$

where the operator Q_l is given by $Q_l=|\varphi_l\rangle\,\langle\boldsymbol{\nabla}\varphi_l|\,\boldsymbol{\nabla}$. In a similar way, we may use $\overline{\langle g_k,\varphi_l\rangle}_{1,2}=\langle\varphi_l,g_k\rangle_{1,2}$ to instead write the spectral weights in equation (22) as $m_{jk}(l)=\langle\boldsymbol{\nabla} Q_l\,g_j|\boldsymbol{\nabla} g_k\rangle$, hence Q_l is a symmetric operator with respect to the inner-product $\langle\cdot,\cdot\rangle_{1,2}$. Since $\langle\boldsymbol{\nabla}\varphi_i|\boldsymbol{\nabla}\varphi_l\rangle=\delta_{il}$ it is clear that the Q_l are mutually orthogonal projection operators $Q_iQ_l=\delta_{il}Q_i$. With this notation, we may formally identify the self-adjoint projection operator $Q(\lambda)$ and the spectral measure $\mathrm{d}\mu_{jk}(\lambda)=\mathrm{d}\langle\boldsymbol{\nabla} Q(\lambda)g_j|\boldsymbol{\nabla} g_k\rangle$ as

$$(23)\quad Q(\lambda)=\sum_{l\,:\,\lambda_l\leq\lambda}\theta(\lambda-\lambda_l)\,Q_l, \quad \mathrm{d}\mu_{jk}(\lambda)=\sum_{l\,:\,\lambda_l\leq\lambda}\delta_{\lambda_l}(\mathrm{d}\lambda)\langle\boldsymbol{\nabla} Q_l\,g_j|\boldsymbol{\nabla} g_k\rangle,$$

where $\theta(\lambda)$ is the Heaviside function and $\delta_{\lambda_l}(\mathrm{d}\lambda)$ is the Dirac δ-measure concentrated at λ_l. This formula is *precisely* true for the matrix setting, where the Q_l are given by mutually orthogonal projection matrices, $\boldsymbol{\nabla}$ is given by a finite difference matrix, and g_j is a Euclidean vector [71].

A key feature of equations (18) and (19) is that parameter information in ε is *separated* from the complicated geometry and dynamics of the time-dependent flow, which are encoded in the spectral measure μ_{jk}. This important property of the integrals in (18) follows from the non-dimensionalization of the advection-diffusion equation discussed in the paragraph leading to equation (3), yielding a spectral measure μ_{jk} that is *independent* of the molecular diffusivity ε. An alternate formulation of the effective parameter problem for advection-diffusion by time-dependent flows was discussed in [6], which used a different non-dimensionalization, yielding a Stieltjes integral representation for S^*_{kk} involving the Péclet number Pe of the flow and a spectral measure that depends on the Strouhal number. However, as pointed out in [16], the Strouhal number dependence of the measure led to an implicit dependence of the spectral measure on Pe. This restricts the utility of the integral representations, such as rigorous bounds [7, 38] which depend explicitly on Pe but also implicitly on Pe through the moments of the measure. Our formulation has no such restrictions.

3. Fourier methods

In equation (19) we provided series representations for the discrete parts of the integral representations for S^*_{jk} and A^*_{jk} shown in (18). These series involve the *real* eigenvalues λ_l, $l = 0, 1, 2, \ldots$, and orthonormal eigenfunctions φ_l of the self-adjoint operator $M = -\imath A$, where $M\varphi_l = \lambda_l \varphi_l$, $\langle \varphi_i, \varphi_l \rangle_{1,2} = \delta_{il}$, and $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})$. In Appendix E.2 we provide a Fourier representation of the eigenvalue problem $M\varphi_l = \lambda_l \varphi_l$, transforming it to an infinite system of algebraic equations involving the trigonometric Fourier coefficients of the φ_l. In this section we refine the mathematical framework, applying it to the fluid velocity field in equation (1). In Section 4 we truncate the resultant infinite system of algebraic equations and write the truncated system as a generalized eigenvalue problem involving symmetric matrices. We then compute the effective diffusivity directly in terms of the eigenvalues and eigenvectors of this generalized eigenvalue problem.

In Appendix E.2 we demonstrate that a Fourier representation of the eigenvalue problem $M\varphi_l = \lambda_l \varphi_l$ follows from expanding the eigenfunctions φ_l and the components u_j, $j = 1, \ldots, d$, of the fluid velocity field $\boldsymbol{u}$ in a trigonometric Fourier series,

$$(24) \quad \varphi_l = \sum_{\ell, \boldsymbol{k}} a^l_{\ell, \boldsymbol{k}} \, \phi_{\ell, \boldsymbol{k}} \,, \quad u_j = \sum_{\ell', \boldsymbol{k}'} b^j_{\ell', \boldsymbol{k}'} \, \phi_{\ell', \boldsymbol{k}'} \,, \qquad \phi_{\ell, \boldsymbol{k}}(t, \boldsymbol{x}) = \mathrm{e}^{\imath(\ell t + \boldsymbol{k}\boldsymbol{\cdot}\boldsymbol{x})},$$

where the series for u_j involves only a *finite* number of terms. Here, $a^l_{\ell,\boldsymbol{k}} = \langle \varphi_l, \phi_{\ell,\boldsymbol{k}}\rangle$, $b^j_{\ell',\boldsymbol{k}'} = \langle u_j, \phi_{\ell',\boldsymbol{k}'}\rangle$, the sesquilinear inner-product $\langle\cdot,\cdot\rangle$ is given by $\langle f, h\rangle = \langle f\,\overline{h}\rangle$, and $\langle\cdot\rangle$ denotes space-time averaging over the period cell $\mathcal{T}\times\mathcal{V}$. In Appendix C.1.2 we show it is necessary that the eigenfunction φ_l in (24) satisfies $\langle\varphi_l\rangle_{\mathcal{V}} = 0$, where $\langle\cdot\rangle_{\mathcal{V}}$ denotes spatial average over $\mathcal{V}$. Therefore, in general, the series for φ_l in equation (24) runs over the index set $I = \{(\ell,\boldsymbol{k}) \in \mathbb{Z}^{d+1} \mid \boldsymbol{k} \neq 0\}$. It is also necessary that $\langle u_j\rangle_{\mathcal{V}} = 0$, though $\langle u_j\rangle_{\mathcal{T}} = 0$ is also allowed but not necessary, where $\langle\cdot\rangle_{\mathcal{T}}$ denotes spatial average over $\mathcal{T}$ (see Appendix C.1.2 for details). The fluid velocity field $\boldsymbol{u}$ in equation (1) satisfies $\langle\boldsymbol{u}\rangle_{\mathcal{V}} = 0$ and $\langle\boldsymbol{u}\rangle = 0$ but $\langle\boldsymbol{u}\rangle_{\mathcal{T}} = (\cos y, \cos x) \not\equiv 0$, and the associated series for u_j in equation (24) runs over the index set $\ell, k_i \in \{-1,0,1\}$, $i = 1,2$, with $\boldsymbol{k} \neq 0$.

In Appendix E.2 we show that inserting the representations for φ_l and u_j in equation (24) into the eigenvalue problem $M\varphi_l = \lambda_l\varphi_l$ and denoting $\boldsymbol{b}_{\ell',\boldsymbol{k}'} = \left(b^1_{\ell',\boldsymbol{k}'}, \ldots, b^d_{\ell',\boldsymbol{k}'}\right)$ yields the Fourier representation of $M\varphi_l = \lambda_l\varphi_l$,

$$|\boldsymbol{k}|^{-2}\Big(\ell\, a^l_{\ell,\boldsymbol{k}} - \sum_{\ell',\boldsymbol{k}'} [\overline{\boldsymbol{b}_{\ell',\boldsymbol{k}'}}\boldsymbol{\cdot}\boldsymbol{k}]\, a^l_{\ell+\ell',\boldsymbol{k}+\boldsymbol{k}'}\Big) = \lambda_l\, a^l_{\ell,\boldsymbol{k}}. \tag{25}$$

Equation (25) is an infinite system of algebraic equations that determines the eigenvalues λ_l and Fourier coefficients $a^l_{\ell,\boldsymbol{k}}$ of the eigenfunctions φ_l of the self-adjoint operator $M = -\imath A$. The Fourier representation of the spectral weights $m_{jk}(l) = \langle u_j, \varphi_l\rangle\,\overline{\langle u_k, \varphi_l\rangle}$ in equation (20) are determined by

$$\langle u_j, \varphi_l\rangle = \sum_{\ell',\boldsymbol{k}'} b^j_{\ell',\boldsymbol{k}'}\,\overline{a^l_{\ell',\boldsymbol{k}'}}\,. \tag{26}$$

We now apply the results shown in equations (25) and (26) to the fluid velocity field $\boldsymbol{u}$ shown in equation (1). In particular, writing $\boldsymbol{u} = (u_1, u_2)$ and $\boldsymbol{x} = (x, y)$ we have

$$u_1(t,x,y) = \cos y + \theta\,\cos t\,\sin y, \qquad \theta \in (0,1], \tag{27}$$

and $u_2(t,x,y) = u_1(t,y,x)$. Using, for example, $\cos t = (\exp(\imath t) + \exp(-\imath t))/2$ and $\sin y = (\exp(\imath y) - \exp(-\imath y))/(2\imath)$, we have

$$u_1(t,x,y) = \frac{1}{2}(\mathrm{e}^{\imath y} + \mathrm{e}^{-\imath y}) + \frac{\theta}{4\imath}(\mathrm{e}^{\imath(t+y)} - \mathrm{e}^{\imath(t-y)} + \mathrm{e}^{\imath(-t+y)} - \mathrm{e}^{\imath(-t-y)}), \tag{28}$$

and similarly for $u_2(t,x,y) = u_1(t,y,x)$. Consequently, denoting $\boldsymbol{k} = (m,n)$, equation (25) can be written as

(29)
$$\begin{aligned}&\frac{\ell\, a^l_{\ell,m,n}}{m^2+n^2} - \frac{1}{m^2+n^2}\Bigg[\frac{1}{2}\Big[m\Big(a^l_{\ell,m,n+1} + a^l_{\ell,m,n-1}) + n(a^l_{\ell,m+1,n} + a^l_{\ell,m-1,n}\Big)\Big]\\ &\quad -\frac{\theta}{4\imath}\Big[m\Big(a^l_{\ell+1,m,n+1} - a^l_{\ell+1,m,n-1} + a^l_{\ell-1,m,n+1} - a^l_{\ell-1,m,n-1}\Big)\\ &\qquad +n\Big(a^l_{\ell+1,m+1,n} - a^l_{\ell+1,m-1,n} + a^l_{\ell-1,m+1,n} - a^l_{\ell-1,m-1,n}\Big)\Big]\Bigg]\\ &\qquad\quad = \lambda_l a^l_{\ell,m,n}, \qquad (m,n)\neq(0,0).\end{aligned}$$

Equations (26) and (28) imply the spectral weights $m_{jk}(l) = \langle u_j, \varphi_l\rangle\,\overline{\langle u_k, \varphi_l\rangle}$ in (20) are determined by

(30)
$$\begin{aligned}\overline{\langle u_1, \varphi_l\rangle} &= \frac{1}{2}\left(a^l_{0,0,1} + a^l_{0,0,-1}\right) - \frac{\theta}{4\imath}\left(a^l_{1,0,1} - a^l_{1,0,-1} + a^l_{-1,0,1} - a^l_{-1,0,-1}\right),\\ \overline{\langle u_2, \varphi_l\rangle} &= \frac{1}{2}\left(a^l_{0,1,0} + a^l_{0,-1,0}\right) - \frac{\theta}{4\imath}\left(a^l_{1,1,0} - a^l_{1,-1,0} + a^l_{-1,1,0} - a^l_{-1,-1,0}\right).\end{aligned}$$

Equation (30) shows, for the flow in equation (1), using the orthonormal trigonometric basis functions $\phi_{\ell,\boldsymbol{k}}(t,\boldsymbol{x}) = \exp[\imath(\ell t + \boldsymbol{k}\cdot\boldsymbol{x})]$ leads to an exact representation of the spectral measure weights $m_{jk}(l) = \langle u_j, \varphi_l\rangle\,\overline{\langle u_k, \varphi_l\rangle}$ which involves a multiplication of two series with only *six* terms. Of course, we could have used a different orthonormal basis. However, the representations of u_1 and u_2 in equation (28) and the spectral weights in equation (30) would then be given by infinite series.

When $\theta = 0$ in equation (1), the fluid velocity field $\boldsymbol{u}$ is time-independent, $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$, the operator A no longer involves the time derivative, and the associated eigenfunction φ_l is also time-independent, $\varphi_l = \varphi_l(\boldsymbol{x})$. In this case, the system of equations in (29) reduces to

$$\text{(31)}\quad \frac{-1}{2(m^2+n^2)}\left[m(a^l_{m,n+1} + a^l_{m,n-1}) + n(a^l_{m+1,n} + a^l_{m-1,n})\right] = \lambda_l a^l_{m,n},$$

where $(m,n)\neq 0$, while equation (30) reduces to

$$\text{(32)}\qquad \overline{\langle u_1, \varphi_l\rangle} = \frac{1}{2}\left(a^l_{0,1} + a^l_{0,-1}\right), \quad \overline{\langle u_2, \varphi_l\rangle} = \frac{1}{2}\left(a^l_{1,0} + a^l_{-1,0}\right).$$

In the following section we will use equations (29)–(32) to compute the discrete parts of the effective diffusivity tensor D^* associated with the flow in equation (1).

4. Numerical results

In equation (19) we provided a series representation for the discrete component $\tilde{\mathsf{S}}^*$ of the symmetric part S^* of the effective diffusivity tensor D^*. This series involves the real eigenvalues λ_l and the orthonormal eigenvectors φ_l of the self-adjoint operator $M = -\imath A$ through the spectral measure weights $m_{jk}(l) = \langle u_j, \varphi_l\rangle \overline{\langle u_k, \varphi_l\rangle}$, which involve the components u_j, $j = 1, \ldots, d$ of the fluid velocity field $\boldsymbol{u}$. In Section 3, we used Fourier methods to transform the eigenvalue problem $M\varphi_l = \lambda_l\varphi_l$ associated with the flow in equation (1), for both $\theta \neq 0$ and $\theta = 0$, into infinite systems of algebraic equations shown in (29) and (31), respectively, involving the trigonometric Fourier coefficients of the eigenfunctions φ_l. We also determined in equations (30) and (32) the spectral weights $m_{jk}(l)$ associated with the fluid velocity field $\boldsymbol{u}$ in equation (1) for $\theta \neq 0$ and $\theta = 0$, respectively. In this section, we truncate these infinite systems, convert them to matrix eigenvalue problems, and numerically compute $\tilde{\mathsf{S}}^*_{kk}$ by directly computing the eigenvalues λ_l and spectral measure weights $m_{jk}(l)$.

By restricting the indices, $-N \leq \ell, m, n \leq N$, and imposing the truncation conditions

$$a^l_{\ell,m,n} = 0 \quad \text{if} \quad \max(|\ell|, |m|, |n|) > N, \tag{33}$$

the infinite systems of equations in (29) and (31) become finite sets of equations. Consider the fluid velocity field in (1) with parameter $\theta \in [0, 1]$. In the dynamic ($\theta \neq 0$) and steady ($\theta = 0$) cases, the bijective mappings $\Theta_d(\ell, m, n)$ and $\Theta_s(m, n)$ defined by

$$\begin{aligned} \Theta_d(\ell, m, n) &= (N + m + 1) + (N + n)(2N + 1) + (N + \ell)(2N + 1)^2, \\ \Theta_s(m, n) &= (N + m + 1) + (N + n)(2N + 1), \end{aligned} \tag{34}$$

map each finite set of equations to a matrix equation $\mathsf{C}^{-1}\mathsf{B}\boldsymbol{a}_l = \lambda_l\boldsymbol{a}_l$ which can be written as the generalized eigenvalue problem

$$\mathsf{B}\boldsymbol{a}_l = \lambda_l\mathsf{C}\boldsymbol{a}_l. \tag{35}$$

Here, B and C is a symmetric and diagonal matrix, respectively. More specifically, B is Hermitian in the dynamic case and is real-symmetric in the steady case. The matrix C is real-symmetric and diagonal in both cases, with the values $|\boldsymbol{k}|^2 = m^2 + n^2$ along its diagonal. Since the indices (m, n) are restricted to $(m, n) \neq 0$, the matrix C is strictly positive definite.

Since B and C are symmetric matrices and C is strictly positive definite, the generalized eigenvalues λ_l are real-valued and the eigenvectors $\boldsymbol{a}_l$ – consisting of the Fourier coefficients for φ_l – satisfy the orthogonality condition [78]

$$\boldsymbol{a}_j \cdot \mathsf{C} \boldsymbol{a}_k = \delta_{jk}. \tag{36}$$

Moreover, since the matrix C is strictly positive definite, the generalized eigenvalue problem in equation (35) can be written as the following standard eigenvalue problem

$$\mathsf{C}^{-1/2} \mathsf{B} \mathsf{C}^{-1/2} \boldsymbol{v}_l = \lambda_l \boldsymbol{v}_l, \qquad \boldsymbol{v}_l = \mathsf{C}^{1/2} \boldsymbol{a}_l. \tag{37}$$

Since B is a symmetric matrix and C is diagonal, the matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$ is also symmetric with real-valued eigenvalues and orthonormal eigenvectors. From the orthogonality relation $\boldsymbol{v}_j \cdot \boldsymbol{v}_k = \delta_{jk}$ we recover equation (36) via $\boldsymbol{v}_l = \mathsf{C}^{1/2} \boldsymbol{a}_l$ in (37).

In summary, our numerical method is the following. Create the matrices B and C according to equation (29) or (31) and the corresponding bijective mapping in (34). Compute *all* of the eigenvalues λ_l and eigenvectors $\boldsymbol{v}_l$ of the symmetric matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$. The computed Fourier coefficients of the eigenfunction φ_l are given by $\boldsymbol{a}_l = \mathsf{C}^{-1/2}\boldsymbol{v}_l$. The eigenvalues λ_l associated with the discrete component of the spectral measure shown in equation (19) are given by the eigenvalues of the matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$, while the spectral measure weights $m_{jk}(l) = \langle u_j, \varphi_l \rangle \overline{\langle u_k, \varphi_l \rangle}$ in (20) are determined from the vector $\boldsymbol{a}_l$ via equation (30) or (32).

In our computations, we used for the steady case $N = 150$, yielding matrices of size $(2N+1)^2 - 1 = 90{,}600$, while in the dynamic case we used $N = 20$, yielding matrices of size $(2N+1)[(2N+1)^2 - 1] = 68{,}880$. The eigenvalues and eigenvectors of the symmetric matrix $\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}$ were computed using the Matlab function *eig()* and used to compute the discrete spectral measure and effective diffusivity as described above. The stability of the computations are measured in terms of the condition numbers $\mathcal{K}_l$ of the eigenvalues λ_l, which are the reciprocals of the cosines of the angles between the left and right eigenvectors. Eigenvalue condition numbers close to 1 indicate a stable computation. Our eigenvalue computations are extremely stable with $\max_l |1 - \mathcal{K}_l| \sim 10^{-14}$, which were computed using the Matlab function *condeig()*.

Displayed in Fig. 1 are our computations of the discrete component of the spectral measure $\mathrm{d}\mu_{11}(\lambda)=\sum_l m_{11}(l)\delta_{\lambda_l}(\mathrm{d}\lambda)$ associated with the fluid velocity field $\boldsymbol{u}$ shown in equation (1), for (a) the steady ($\theta=0$) and (b) the dynamic ($\theta=1$) settings. Here, the spectral weights $m_{11}(l)=|\langle u_1,\varphi_l\rangle|^2$ are determined by equations (32) and (30), respectively. Consistent with the symmetries of the flows [14], we have $\mu_{11}=\mu_{22}$, while $\mathrm{Re}\,\mu_{12}=0$ and $\mathrm{Im}\,\mu_{12}=0$, up to numerical accuracy and finite size effects.

For the 2D steady cell flow in (1) with $\theta=0$, it is known [29, 74] that $\mathsf{S}^*_{11}\sim\varepsilon^{1/2}$ for $\varepsilon\ll 1$. Our computation of S^*_{11} displayed in Fig. 1(c) is in excellent agreement with this result, with a computed critical exponent of ≈ 0.52 having an error of only 4% relative to its true value 0.5. Reducing N from 150 to 100 changes the value of the critical exponent by less than 0.0015, indicating that the value of $N=150$ is sufficiently large. In this steady setting, the underlying operator $(-\Delta)^{-1}[\boldsymbol{u}_1\cdot\boldsymbol{\nabla}]$ is compact [12] and therefore has bounded, discrete spectrum away from the spectral origin, with a limit point at $\lambda=0$ [95]. The limit point behavior of the measure μ_{11} can be seen in the rightmost panel of Fig. 1(a). The decay of S^*_{11} for vanishing ε is due to the magnitude of the measure masses $m_{11}(l)\lesssim 10^{-30}$ for $|\lambda_l|\ll 1$, with a significant *spectral gap* near the limit point. The rigorous result [29, 74] $\mathsf{S}^*_{11}\sim\varepsilon^{1/2}$ as $\varepsilon\to 0$ reveals that the spectrum of the operator $(-\Delta)^{-1}[\boldsymbol{u}_1\cdot\boldsymbol{\nabla}]$ at $\lambda=0$ is either continuous or it is discrete with zero mass, otherwise S^*_{11} would diverge as $\varepsilon\to 0$.

In contrast, as shown in Fig. 1(b), the spectral measure μ_{11} associated with the time-dependent fluid velocity field in (1), with $\theta=1$, has significant values of $m_{11}(l)$ near the spectral origin, with $m_{11}(l)\gtrsim 10^{-10}$ more than *20 orders* of magnitude greater than that of the steady flow. A limit point behavior in the measure μ_{11} near $\lambda=0$ can be seen in the rightmost panel of Fig. 1(b). It is interesting to note that the support *supp* μ_{11} of the measure μ_{11} increases with N and satisfies *supp* $\mu_{11}\subset[-N,N]$ for all values of N investigated, which suggests that *supp* μ_{11} becomes an unbounded set as $N\to\infty$. This is consistent with the unboundedness of the self-adjoint operator $M=-\imath(-\Delta)^{-1}(\partial_t-\boldsymbol{u}\cdot\boldsymbol{\nabla})$. Due to the significant mass of the measure near the spectral origin and its uniform nature, as shown in the center panel of Fig. 1(b), the effective diffusivity has an $O(1)$ behavior, $\mathsf{S}^*_{11}\sim 1$ for $\varepsilon\ll 1$, as shown in Fig. 1(d). This is consistent with numerical computations of S^*_{11} using alternate methods [14]. This $O(1)$ behavior of S^*_{11} has been attributed to Lagrangian chaos exhibited by the flow in (1) [14, 110]. This phenomenon is called *residual diffusion* since the chaotic mixing of the flow gives rise to large scale macroscopic transport even in the absence of molecular diffusion, $\varepsilon\to 0$.

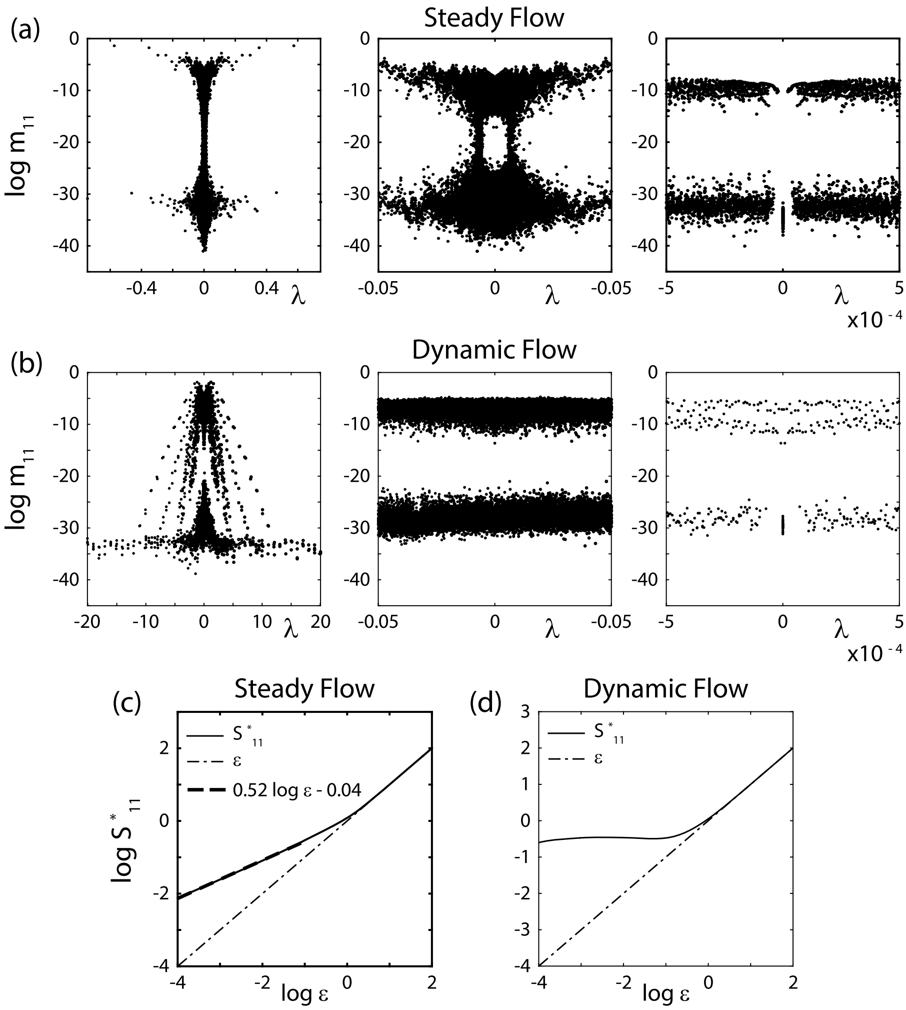

Figure 1: Computations of spectral measures and effective diffusivities for steady and dynamic flows. The spectral measure μ_{11} associated with the flow in (1) are displayed with increasing λ–axis magnifications from left to right for (a) the steady setting and (b) the dynamic setting with the associated effective diffusivity S^*_{11} displayed in (c) and (d), respectively. In the steady case (a), the limit point of the measure near $\lambda = 0$ has small measure mass with $m_{11} \lesssim 10^{-30}$, leading to the asymptotic behavior $\mathsf{S}^*_{11} \sim \varepsilon^{1/2}$ for $\varepsilon \ll 1$, displayed in (c). In the dynamic case (b), the significant measure mass $m_{11} \gtrsim 10^{-10}$ near $\lambda = 0$ leads to the asymptotic behavior $\mathsf{S}^*_{11} \sim 1$ for $\varepsilon \ll 1$, displayed in (d).

5. Conclusions

We have adapted and extended two approaches of the effective parameter problem for advection enhanced diffusion proposed in [4, 5] and [79], from the setting of a time-independent fluid velocity field $\boldsymbol{u}$ to space-time periodic $\boldsymbol{u}$, allowing for the possibility of chaotic dynamics. For each approach, we formulated a rigorous mathematical framework which provides Stieltjes integral representations for both the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* for such flows, involving a spectral measure of an *unbounded* self-adjoint operator. We also used abstract methods of functional analysis to prove that the two approaches produce equivalent spectral representations for D^*. The approach proposed in [4, 5] is based on the analytic continuation method for representing transport in composite materials [38], though the definitions of the effective parameters are different. We generalized a result in [29] to the time-dependent setting, providing a precise relationship between D^* and the effective parameter arising from the definition used in the analytic continuation method.

The integral representations for D^* involve a Stieltjes measure that has continuous and discrete components. We have provided a mathematical foundation for rigorous computation of the discrete part of D^*, which involves the eigenvalues and eigenfunctions of the associated self-adjoint, integro-differential operator. In particular, we developed Fourier methods to represent the eigenvalue problem as an infinite system of algebraic equations involving the trigonometric Fourier coefficients of the eigenfunctions. We truncated this system of equations to obtain a generalized eigenvalue problem involving symmetric matrices. The discrete part of the spectral measure and D^* are given explicitly in terms of the generalized eigenvalues and eigenvectors of the matrices. We implemented this method to compute D^* for a model cell flow and a time-dependent flow exhibiting Lagrangian chaos. Our Fourier approach has accurately captured the known asymptotic behavior of the the cell flow in the advection dominated regime. Our approach has also captured the phenomenon known as residual diffusion related to the Lagrangian chaos of the flow, where chaotic mixing of the flow gives rise to large scale macroscopic transport even in the absence of molecular diffusion.

Appendix A. Spectral theory of unbounded self-adjoint operators in Hilbert space

The theory of *unbounded* operators in Hilbert space was developed largely by John von Neumann and Marshall H. Stone. It is considerably more technical and challenging than the theory of bounded operators, as unbounded operators do not form an algebra, nor even a linear space, because each one is defined on its own domain. In this section, we review the spectral theory for such operators and, in particular, the celebrated *spectral theorem* for self-adjoint operators [88, 97].

An operator is not determined unless its domain is known. Let Φ_1 and Φ_2 be operators acting on a Hilbert space $\mathscr{H}$ with domains $D(\Phi_1)$ and $D(\Phi_2)$, respectively, $D(\Phi_i) \subset \mathscr{H}$, $i = 1, 2$. They are said to be *identical*, in symbols $\Phi_1 \equiv \Phi_2$, if and only if $D(\Phi_1) = D(\Phi_2)$ and $\Phi_1 f = \Phi_2 f$ for every f of their common domain. They are said to be *equal* in the set $\mathscr{S}$, in symbols $\Phi_1 = \Phi_2$, if and only if $\mathscr{S} \subseteq D(\Phi_1) \cap D(\Phi_2)$ and $\Phi_1 f = \Phi_2 f$ for every $f \in \mathscr{S}$. The operator Φ_2 is said to be an *extension* (*proper extension*) of the operator Φ_1 if $D(\Phi_1) \subseteq D(\Phi_2)$ ($D(\Phi_1) \subset D(\Phi_2)$) and the operators Φ_2 and Φ_1 are equal in $D(\Phi_1)$ [97].

Consider the sesquilinear inner-product $\langle \cdot, \cdot \rangle$ associated with $\mathscr{H}$ satisfying $\langle a\psi, b\varphi \rangle = a\,\overline{b}\,\langle \psi, \varphi \rangle$ and $\langle \psi, \varphi \rangle = \overline{\langle \varphi, \psi \rangle}$ for all $\psi, \varphi \in \mathscr{H}$ and $a, b \in \mathbb{C}$, where $\overline{z}$ denotes complex conjugation of $z \in \mathbb{C}$. The $\mathscr{H}$–inner-product induces a norm $\| \cdot \|$ defined by $\|\psi\| = \langle \psi, \psi \rangle^{1/2}$. A linear operator Φ is said to be *closed* if for every pair of sequences $\{f_n\}$ and $\{\Phi f_n\}$ (with $f_n \in D(\Phi)$) that converge in the norm $\| \cdot \|$ to the limits f and h, then these limits satisfy $f \in D(\Phi)$ and $\Phi f = h$ [97]. The (Hilbert space) adjoint Φ^* of Φ is defined by $\langle \Phi\psi, \varphi \rangle = \langle \psi, \Phi^* \varphi \rangle$ for every $\psi \in D(\Phi)$ and $\varphi \in D(\Phi^*)$. The adjoint Φ^* of Φ is uniquely determined when the domain $D(\Phi)$ *determines* $\mathscr{H}$, i.e., the smallest closed linear manifold containing $D(\Phi)$ is the Hilbert space $\mathscr{H}$ [97]. In this case, $D(\Phi) \subseteq D(\Phi^*)$ and Φ^* is a closed linear operator [97]. The operator Φ is said to be *symmetric* if $\Phi = \Phi^*$. The operator Φ is said to be *self-adjoint* if $\Phi \equiv \Phi^*$. A symmetric operator is said to be *maximal* if it has no proper symmetric extension. A self-adjoint operator is a maximal symmetric operator [97].

The operator Φ is said to be *bounded* (in operator norm) if $\|\Phi\| = \sup_{\{\psi \in \mathscr{H} \,:\, \|\psi\| = 1\}} \|\Phi\psi\| < \infty$. A bounded linear symmetric operator is self-adjoint if and only if its domain is $\mathscr{H}$ [97]. In particular, the Hellinger–Toeplitz theorem states that, if the operator Φ satisfies $\langle \Phi\psi, \varphi \rangle = \langle \psi, \Phi\varphi \rangle$ for *every* $\psi, \varphi \in \mathscr{H}$, then Φ is bounded on $\mathscr{H}$ [88]. This indicates that, if Φ is an *unbounded* symmetric operator on $\mathscr{H}$, then it is self-adjoint only on a *proper subset* of $\mathscr{H}$ that is dense in $\mathscr{H}$ [88, 97].

The spectrum Σ of a self-adjoint operator Φ on a Hilbert space $\mathscr{H}$ is real-valued [88, 97]. If Φ is also bounded, then its spectral radius equal to its operator norm $\|\Phi\|$ [88], i.e.,

$$\Sigma \subseteq [-\|\Phi\|, \|\Phi\|\,]. \tag{A-2}$$

If Φ is instead unbounded, its spectrum Σ can be an unbounded subset of, or can even coincide with the set of real numbers $\mathbb{R}$ [97].

We now summarize the spectral theorem for self-adjoint operators (see Theorems 5.9 and 6.1 in [97]). Let Φ be a fixed self-adjoint operator with spectrum $\Sigma \subseteq \mathbb{R}$ and domain $D(\Phi)$ that is dense in $\mathscr{H}$. If Φ is bounded then we simply take $D(\Phi) \equiv \mathscr{H}$. The spectral theorem states that there is a one-to-one correspondence between the self-adjoint operator Φ and a family of self-adjoint projection operators $\{Q(\lambda)\}_{\lambda\in\Sigma}$ — the resolution of the identity — that satisfies [97]

$$\lim_{\lambda\to\inf\Sigma} Q(\lambda)=0, \qquad \lim_{\lambda\to\sup\Sigma} Q(\lambda)=I, \tag{A-3}$$

where 0 and I denote the null and identity operators on $\mathscr{H}$, respectively. Furthermore, the *complex-valued* function of the spectral variable λ defined by $\mu_{\psi\varphi}(\lambda)=\langle Q(\lambda)\psi,\varphi\rangle$ has real, $\operatorname{Re}\mu_{\psi\varphi}(\lambda)$, and imaginary, $\operatorname{Im}\mu_{\psi\varphi}(\lambda)$, parts that are of bounded variation for all $\psi,\varphi\in D(\Phi)$ and $\lambda\in\Sigma$ [97], where

$$\begin{aligned}\operatorname{Re}\mu_{\psi\varphi}(\lambda)&=\frac{1}{2}\left(\mu_{\psi\varphi}(\lambda)+\overline{\mu}_{\psi\varphi}(\lambda)\right),\\ \operatorname{Im}\mu_{\psi\varphi}(\lambda)&=\frac{1}{2\imath}\left(\mu_{\psi\varphi}(\lambda)-\overline{\mu}_{\psi\varphi}(\lambda)\right),\end{aligned} \tag{A-4}$$

$\imath=\sqrt{-1}$, and $\lambda\in\Sigma$.

By the sesquilinearity of the inner-product and the fact that the projection operator $Q(\lambda)$ is self-adjoint, the function $\mu_{\psi\varphi}(\lambda)$ satisfies $\mu_{\varphi\psi}(\lambda)=\overline{\mu}_{\psi\varphi}(\lambda)$. Moreover, the function $\mu_{\psi\psi}(\lambda)$ is real-valued and positive, $\mu_{\psi\psi}(\lambda)=\langle Q(\lambda)\psi,\psi\rangle=\langle Q(\lambda)\psi,Q(\lambda)\psi\rangle=\|Q(\lambda)\psi\|^2\geq 0$, hence $\operatorname{Re}\mu_{\psi\psi}(\lambda)=\mu_{\psi\psi}(\lambda)$ and $\operatorname{Im}\mu_{\psi\psi}(\lambda)=0$. With each of these functions of bounded variation, we associate Stieltjes measures [96, 97, 34]

$$\begin{aligned}\mathrm{d}\mu_{\psi\varphi}(\lambda)&=\mathrm{d}\langle Q(\lambda)\psi,\varphi\rangle, & \mathrm{dRe}\,\mu_{\psi\varphi}(\lambda)&=\mathrm{dRe}\,\langle Q(\lambda)\psi,\varphi\rangle,\\ \mathrm{d}\mu_{\psi\psi}(\lambda)&=\mathrm{d}\|Q(\lambda)\psi\|^2, & \mathrm{dIm}\,\mu_{\psi\varphi}(\lambda)&=\mathrm{dIm}\,\langle Q(\lambda)\psi,\varphi\rangle,\end{aligned} \tag{A-5}$$

which we will denote by $\mu_{\psi\psi}$, $\mu_{\psi\varphi}$, $\operatorname{Re}\mu_{\psi\varphi}$, and $\operatorname{Im}\mu_{\psi\varphi}$. We stress that $\mu_{\psi\psi}$ is a positive measure, $\mu_{\psi\varphi}$ is a complex measure, while $\operatorname{Re}\mu_{\psi\varphi}$ and $\operatorname{Im}\mu_{\psi\varphi}$ are signed measures [96, 97].

The spectral theorem also provides an operational calculus in Hilbert space which yields powerful integral representations involving the Stieltjes measures shown in equation (A-5). A summary of the relevant details is as follows. Let $F(\lambda)$ and $G(\lambda)$ be arbitrary complex-valued functions and denote by $\mathscr{D}(F)$ the set of all $\psi \in D(\Phi)$ such that $F \in L^2(\mu_{\psi\psi})$, i.e., F is square integrable on the set Σ with respect to the *positive* measure $\mu_{\psi\psi}$, and similarly define $\mathscr{D}(G)$. Then $\mathscr{D}(F)$ and $\mathscr{D}(G)$ are linear manifolds and there exist linear operators denoted by $F(\Phi)$ and $G(\Phi)$ with domains $\mathscr{D}(F)$ and $\mathscr{D}(G)$, respectively, which are defined in terms of the following Radon–Stieltjes integrals [97]

$$\text{(A-6)}\quad \begin{aligned} \langle F(\Phi)\psi, \varphi\rangle &= \int_{-\infty}^{\infty} F(\lambda)\, \mathrm{d}\mu_{\psi\varphi}(\lambda), \qquad && \forall\, \psi \in \mathscr{D}(F),\ \varphi \in \mathscr{H}, \\ \langle F(\Phi)\psi, G(\Phi)\varphi\rangle &= \int_{-\infty}^{\infty} F(\lambda)\overline{G}(\lambda)\, \mathrm{d}\mu_{\psi\varphi}(\lambda), && \forall\, \psi \in \mathscr{D}(F),\ \varphi \in \mathscr{D}(G), \end{aligned}$$

where the integration in (A-6) is over the spectrum Σ of Φ [88, 97].

The mass $\mu^0_{\psi\varphi} = \int_{-\infty}^{\infty} \mathrm{d}\mu_{\psi\varphi}(\lambda)$ of the Stieltjes measure $\mu_{\psi\varphi}$ satisfies [97] $\mu^0_{\psi\varphi} = \lim_{\lambda\to\sup\Sigma} \mu_{\psi\varphi}(\lambda) - \lim_{\lambda\to\inf\Sigma} \mu_{\psi\varphi}(\lambda)$. Consequently, equation (A-3) and the Cauchy–Schwartz inequality yield

$$\text{(A-7)}\qquad \mu^0_{\psi\varphi} = \int_{-\infty}^{\infty} \mathrm{d}\langle Q(\lambda)\psi, \varphi\,\rangle = \langle \psi, \varphi\rangle, \qquad |\mu^0_{\psi\varphi}| \le \|\psi\|\,\|\varphi\| < \infty.$$

Equation (A-7) demonstrates that the measures in (A-5) are *finite measures*, i.e., they have bounded mass [97].

The operators encountered in the ensuing appendices are *skew-adjoint* operators, which are an example of normal operators. Equation (A-6) can be generalized, holding with suitable notational changes, for *maximal normal operators* [97]. Such a normal operator $\mathbf{N}$ with domain $D(\mathbf{N})$ dense in $\mathscr{H}$ commutes with its adjoint $\mathbf{N}^*$, i.e., $\mathbf{N}\mathbf{N}^* = \mathbf{N}^*\mathbf{N}$, and can be decomposed as $\mathbf{N} = \Phi_1 + \imath\Phi_2$, where Φ_1 and Φ_2 are self-adjoint and commute. The spectrum of the normal operator $\mathbf{N}$ is a (possibly unbounded) subset of $\mathbb{C}$ [97]. A special case of a normal operator is a *skew-adjoint* operator satisfying $\mathbf{N}^* = -\mathbf{N}$. It can be decomposed as $\mathbf{N} = \imath\Phi_2$ and since Φ_2 is self-adjoint having purely real spectrum, the skew-adjoint operator $\mathbf{N} = \imath\Phi_2$ has purely imaginary spectrum [97]. Consequently, given such a maximal skew-adjoint operator, one can focus attention on the self-adjoint operator $\Phi_2 = -\imath\mathbf{N}$ without having to resort to the more notationally complicated spectral theory of normal operators.

The signed measures $\operatorname{Re}\mu_{\psi\varphi}$ and $\operatorname{Im}\mu_{\psi\varphi}$ shown in (A-5) arise naturally when considering a maximal skew-adjoint operator $\mathbf{N} = \imath\Phi$, where Φ is self-adjoint. This can be illustrated by considering some special cases that arise naturally in Appendix C below. Consider the functional $\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle$

involving *real-valued* Hilbert space members $F(\mathbf{N})\psi$ and $G(\mathbf{N})\varphi$, so that $\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle = \langle G(\mathbf{N})\varphi, F(\mathbf{N})\psi\rangle \in \mathbb{R}$ and, in particular,

$$\text{(A-8)} \qquad \langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle = \frac{1}{2}(\langle F(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle + \langle G(\mathbf{N})\varphi, F(\mathbf{N})\psi\rangle).$$

Now consider the special cases $F(\mathbf{N}) = G(\mathbf{N})$ and $F(\mathbf{N}) = \mathbf{N}G(\mathbf{N})$, i.e., $F(\imath\lambda) = G(\imath\lambda)$ and $F(\imath\lambda) = \imath\lambda G(\imath\lambda)$ in equation (A-6), respectively. From equations (A-6) and (A-8), the identities $\operatorname{Re} z = (z+\overline{z})/2$ and $\operatorname{Im} z = (z-\overline{z})/(2\imath)$, and the linearity properties [97] of Stieltjes-Radon integrals with respect to the functions $\mu_{\psi\varphi}(\lambda)$ and $\overline{\mu}_{\psi\varphi}(\lambda)$, we have

$$\text{(A-9)} \qquad \begin{aligned} \langle G(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle &= \int_{-\infty}^{\infty} |G(\imath\lambda)|^2 \, \mathrm{dRe}\, \mu_{\psi\varphi}(\lambda), \\ \langle \mathbf{N}G(\mathbf{N})\psi, G(\mathbf{N})\varphi\rangle &= -\int_{-\infty}^{\infty} \lambda\, |G(\imath\lambda)|^2 \, \mathrm{dIm}\, \mu_{\psi\varphi}(\lambda). \end{aligned}$$

An important property of a self-adjoint operator Φ which will be used in Appendix D is that its domain $D(\Phi)$ comprises those and only those elements $\psi \in \mathscr{H}$ such that the Stieltjes integral $\int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}\mu_{\psi\psi}(\lambda)$ is convergent. When $\psi \in D(\Phi)$ the element $\Phi\psi$ is determined by the relations [97]

$$\text{(A-10)} \qquad \langle \Phi\psi, \varphi\rangle = \int_{-\infty}^{\infty} \lambda \, \mathrm{d}\mu_{\psi\varphi}(\lambda), \qquad \|\Phi\psi\|^2 = \int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}\mu_{\psi\psi}(\lambda),$$

where φ is an arbitrary element in $D(\Phi)$ [97]. In fact, this determines the one-to-one correspondence between the self-adjoint operator Φ and its resolution of the identity $Q(\lambda)$ [97].

Appendix B. Time derivative as a maximal normal operator

A key example of an unbounded operator is the time derivative ∂_t acting on the space $L^2(\mathcal{T})$ of Lebesgue measurable functions that are also square integrable on the interval $\mathcal{T} = [0, T]$, say. The unboundedness of ∂_t as an operator on $L^2(\mathcal{T})$ can be understood by considering the orthonormal set of functions $\{\varphi_n\} \subset L^2(\mathcal{T})$ defined by

$$\text{(A-11)} \qquad \varphi_n(t) = \beta \sin(n\pi t/T), \quad \beta = \sqrt{2/T}, \qquad \langle \varphi_n, \varphi_m\rangle_2 = \delta_{nm},$$

where $n, m \in \mathbb{N}$ and $\langle \cdot, \cdot \rangle_2$ denotes the sesquilinear $L^2(\mathcal{T})$–inner-product. It follows from $\partial_t \varphi_n = (n\pi\beta/T)\cos(n\pi t/T)$ and $\|\partial_t \varphi_n\|^2 = (n\pi/T)^2$, that the norm of $\partial_t \varphi_n$ grows arbitrarily large as $n \to \infty$. This clearly demonstrates the unboundedness of the operator ∂_t with domain $L^2(\mathcal{T})$.

When one also imposes periodic or Dirichlet boundary conditions, integration by parts demonstrates that the operator ∂_t is *skew-adjoint* on $L^2(\mathcal{T})$ so that $-\imath\partial_t$ is symmetric with respect to the sesquilinear inner-product $\langle\cdot,\cdot\rangle_2$. We now identify an everywhere dense subset of $L^2(\mathcal{T})$ on which $-\imath\partial_t$ is a linear self-adjoint operator [88, 97]. Consider the class $\mathscr{A}_\mathcal{T}$ of all functions $\psi \in L^2(\mathcal{T})$ such that $\psi(t)$ is *absolutely continuous* [89] on the interval $\mathcal{T}$, having derivative $\psi'(t)$ belonging to $L^2(\mathcal{T})$, i.e., [97, 89]

$$\mathscr{A}_\mathcal{T} = \left\{ \psi \in L^2(\mathcal{T}) \;\middle|\; \psi(t) = c + \int_0^t g(s)ds, \quad g \in L^2(\mathcal{T}) \right\}, \tag{A-12}$$

where the constant c and function $g(s)$ are arbitrary. Now, consider the set $\tilde{\mathscr{A}_\mathcal{T}}$ of all functions $\psi \in \mathscr{A}_\mathcal{T}$ that satisfy the periodic boundary condition $\psi(0) = \psi(T)$, i.e. functions ψ satisfying the properties of equation (A-12) with c arbitrary and $\int_0^T g(s)ds = 0$. In order to help clarify the ideas that were discussed in Appendix A in terms of an abstract Hilbert space $\mathscr{H}$, we also consider the set $\hat{\mathscr{A}_\mathcal{T}}$ of all functions $\psi \in \mathscr{A}_\mathcal{T}$ that satisfy the Dirichlet boundary condition $\psi(0) = \psi(T) = 0$, i.e. functions ψ satisfying the properties of equation (A-12) with $c = 0$ and $\int_0^T g(s)ds = 0$. More concisely,

$$\begin{aligned} \tilde{\mathscr{A}_\mathcal{T}} &= \{\psi \in \mathscr{A}_\mathcal{T} \,|\, \psi(0) = \psi(T)\}, \\ \hat{\mathscr{A}_\mathcal{T}} &= \{\psi \in \mathscr{A}_\mathcal{T} \,|\, \psi(0) = \psi(T) = 0\}. \end{aligned} \tag{A-13}$$

These function spaces satisfy $\hat{\mathscr{A}_\mathcal{T}} \subset \tilde{\mathscr{A}_\mathcal{T}} \subset \mathscr{A}_\mathcal{T}$ and are each everywhere dense in $L^2(\mathcal{T})$ [97]. Let the operators B, $\tilde{B}$, and $\hat{B}$ be identified as $-\imath\partial_t$ with domains $\mathscr{A}_\mathcal{T}$, $\tilde{\mathscr{A}_\mathcal{T}}$, and $\hat{\mathscr{A}_\mathcal{T}}$, respectively. Then, $\hat{B}$ is a closed linear symmetric operator with the adjoint $\hat{B}^* \equiv B$, and the operator $\tilde{B}$ is a *self-adjoint* extension of $\hat{B}$ [97]. In symbols, this means that $\tilde{B} = \tilde{B}^*$ on $\tilde{\mathscr{A}_\mathcal{T}}$ and $D(\tilde{B}) = D(\tilde{B}^*) = \tilde{\mathscr{A}_\mathcal{T}}$, i.e., $\tilde{B} \equiv \tilde{B}^*$ on $\tilde{\mathscr{A}_\mathcal{T}}$. This establishes that the operator $-\imath\partial_t$ with domain $\tilde{\mathscr{A}_\mathcal{T}}$ is self-adjoint, hence ∂_t is a maximal skew-adjoint (normal) operator on $\tilde{\mathscr{A}_\mathcal{T}}$ [97]. The operator $\imath\partial_t$ on $\tilde{\mathscr{A}_\mathcal{T}}$ has a simple point spectrum, consisting of eigenvalues $\lambda = 2n\pi/T$, $n \in \mathbb{Z}$, with corresponding eigenfunctions $\exp(\imath\, 2n\pi t/T)$ [97].

Appendix C. Hilbert spaces, resolvents, and integral representations of the effective diffusivity

In this section we provide a spectral theory of effective diffusivities for space-time periodic flows. In particular, we consider two different approaches to the effective parameter problem for advection-diffusion which were proposed in [79, 12] and [4, 5] for *time-independent* flows. We adapt and extend these results to the setting of *time-dependent* flows, allowing fluid velocity fields with chaotic dynamics. Specifically, we formulate rigorous mathematical frameworks for each approach which provide Stieltjes integral representations for both the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* for space-time periodic flows, involving a spectral measure of an *unbounded* self-adjoint operator. In Appendix C.1 we generalize the approach proposed in [79], while in Appendix C.2 we extend the approach proposed in [4, 5]. In Appendix D we establish that the two approaches are equivalent, using the one-to-one correspondence between a self-adjoint operator and its resolution of the identity [97], discussed in the paragraph containing equation (A-10).

C.1. Scalar fields and effective diffusivity

In this section we provide an abstract Hilbert space formulation of the effective parameter problem for advection-diffusion that was proposed in [79], based on [12], generalizing it to the setting of a space-time periodic fluid velocity field, allowing for flows with chaotic dynamics. To fix ideas, consider the following sets $\mathcal{T}=[0,T]$ and $\mathcal{V}=\otimes_{j=1}^{d}[0,L]$ which define the space-time period cell $\mathcal{T}\times\mathcal{V}$. Now consider the Hilbert spaces $L^2(\mathcal{T})$ and $L^2(\mathcal{V})$ of Lebesgue measurable scalar functions over the complex field $\mathbb{C}$ that are also square integrable [34]. Define the associated Hilbert spaces $\mathscr{H}_{\mathcal{T}}$, $\mathscr{H}_{\mathcal{V}}$, and $\mathscr{H}_{\mathcal{T}\mathcal{V}}=\mathscr{H}_{\mathcal{T}}\otimes\mathscr{H}_{\mathcal{V}}$ of periodic functions, where

$$
\begin{aligned}
\mathscr{H}_{\mathcal{T}} &= \big\{\psi\in L^2(\mathcal{T})\,|\,\psi(t)=\psi(t+T)\big\},\\
\mathscr{H}_{\mathcal{V}} &= \big\{\psi\in L^2(\mathcal{V})\,|\,\psi(\boldsymbol{x})=\psi(\boldsymbol{x}+L\boldsymbol{e}_j),\ j=1,\dots,d\big\},
\end{aligned}
\tag{A-14}
$$

and the $\boldsymbol{e}_j$ are standard basis vectors.

More specifically, denote time average over $\mathcal{T}$ by $\langle\cdot\rangle_{\mathcal{T}}$, space average over $\mathcal{V}$ by $\langle\cdot\rangle_{\mathcal{V}}$, and space-time average over $\mathcal{T}\times\mathcal{V}$ by $\langle\cdot\rangle$. The space-time average $\langle\cdot\rangle$, induces a sesquilinear inner-product $\langle\cdot,\cdot\rangle$ given by $\langle\psi,\varphi\rangle=\langle\psi\,\overline{\varphi}\rangle$, with $\langle\varphi,\psi\rangle=\overline{\langle\psi,\varphi\rangle}$. This $\mathscr{H}_{\mathcal{T}\mathcal{V}}$–inner-product, in turn, induces a norm $\|\cdot\|$ given by $\|\psi\|=\langle\psi,\psi\rangle^{1/2}$ [34]. The set of space-time periodic Lebesgue measurable functions $\mathscr{H}_{\mathcal{T}\mathcal{V}}$ satisfying $\|f\|<\infty$ is a (complete) Hilbert space [34]. Similarly, the space and time averages, $\langle\cdot\rangle_{\mathcal{V}}$ and $\langle\cdot\rangle_{\mathcal{T}}$, induce sesquilinear inner-products, $\langle\cdot,\cdot\rangle_{\mathcal{V}}$ and $\langle\cdot,\cdot\rangle_{\mathcal{T}}$, that induce norms, $\|\cdot\|_{\mathcal{V}}$ and $\|\cdot\|_{\mathcal{T}}$, associated with the Hilbert spaces $\mathscr{H}_{\mathcal{V}}$ and $\mathscr{H}_{\mathcal{T}}$.

To treat temporal dependence, we defined in equation (A-13) the space $\tilde{\mathscr{A}}_{\mathcal{T}}$ of absolutely continuous $\mathcal{T}$–periodic functions with time derivatives belonging to $L^2(\mathcal{T})$, which is *not* a Hilbert space but is instead an everywhere dense subset of the Hilbert space $\mathscr{H}_{\mathcal{T}}$ [97]. To treat spatial dependence, we now define the Sobolev space $\mathscr{H}_{\mathcal{V}}^{1,2}$ which is itself a Hilbert space [12, 33, 63],

$$\mathscr{H}_{\mathcal{V}}^{1,2} = \big\{\psi \in \mathscr{H}_{\mathcal{V}} \mid \|\boldsymbol{\nabla}\psi\|_{\mathcal{V}} < \infty,\ \langle\psi\rangle_{\mathcal{V}} = 0\big\}. \tag{A-15}$$

The condition $\langle\psi\rangle_{\mathcal{V}} = 0$ in (A-15) is required to eliminate non-zero constant ψ, which satisfies $\|\boldsymbol{\nabla}\psi\|_{\mathcal{V}} = 0$. The $\mathscr{H}_{\mathcal{V}}^{1,2}$–norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$ is induced by the $\mathscr{H}_{\mathcal{V}}^{1,2}$–inner-product: $\|\boldsymbol{\nabla}\psi\|_{\mathcal{V}} = \langle\boldsymbol{\nabla}\psi\boldsymbol{\cdot}\boldsymbol{\nabla}\psi\rangle_{\mathcal{V}}^{1/2}$. Define $C^k(\mathcal{V})$ to be the space of all $\mathcal{V}$–periodic, mean-zero, k-times continuously differentiable functions in $\mathscr{H}_{\mathcal{V}}$. We define the Sobolev space $\mathscr{H}_{\mathcal{V}}^{1,2}$ to be the closure in the norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$ of the space $C^3(\mathcal{V})$, containing those elements of $\mathscr{H}_{\mathcal{V}}$ that are mean-zero and have square integrable gradients on the set $\mathcal{V}$. Functions in $\mathscr{H}_{\mathcal{V}}^{1,2}$ need not be differentiable in the classical sense. Instead, $f \in \mathscr{H}_{\mathcal{V}}^{1,2}$ has derivatives $\partial f/\partial x_j \in L^2(\mathcal{V})$ defined by $\partial f/\partial x_j = \lim_{n\to\infty} \partial f_n/\partial x_j$, where $f_n \in C^3(\mathcal{V})$ are Cauchy in the norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$, and convergence is in $L^2(\mathcal{V})$ [63]. We specifically chose $\mathscr{H}_{\mathcal{V}}^{1,2}$ to be the completion of $C^3(\mathcal{V})$ so that the Cauchy sequence $\{f_n\}$ associated with $f \in \mathscr{H}_{\mathcal{V}}^{1,2}$ satisfies $\partial f_n/\partial x_j \in C^2(\mathcal{V})$, for all $j = 1, \dots, d$. We will repeatedly use this property of the Hilbert space $\mathscr{H}_{\mathcal{V}}^{1,2}$.

To characterize the spatial dependence of the function u_j on the right side of the cell problem in (9), we define the Hilbert space $\mathscr{H}_{\mathcal{V}}^{0,2}$ to be the closure in the norm $\|\cdot\|_{\mathcal{V}}$ of the space $C^2(\mathcal{V})$. All the elements of $\mathscr{H}_{\mathcal{V}}^{0,2}$ are those elements of $\mathscr{H}_{\mathcal{V}}$ which are limits of sequences $\{f_n\} \subset C^2(\mathcal{V})$ that are Cauchy in the norm $\|\cdot\|_{\mathcal{V}}$.

Finally, define the Hilbert space $\mathscr{H}$ and its everywhere dense subset $\mathscr{F}$

$$\mathscr{H} = \mathscr{H}_{\mathcal{T}} \otimes \mathscr{H}_{\mathcal{V}}^{1,2}, \qquad \mathscr{F} = \tilde{\mathscr{A}}_{\mathcal{T}} \otimes \mathscr{H}_{\mathcal{V}}^{1,2}. \tag{A-16}$$

Due to the presence of $\tilde{\mathscr{A}}_{\mathcal{T}}$ in the definition of the function space $\mathscr{F}$, it is *not* a complete Hilbert space, and is instead an everywhere dense subset of the complete Hilbert space $\mathscr{H}$. Recall that $\langle\cdot\rangle$ denotes space-time average over $\mathcal{T} \times \mathcal{V}$ and $\boldsymbol{\xi}\boldsymbol{\cdot}\boldsymbol{\zeta} = \boldsymbol{\xi}^T\,\overline{\boldsymbol{\zeta}}$. The sesquilinear $\mathscr{H}$–inner-product is given by $\langle\psi,\varphi\rangle_{1,2} = \langle\boldsymbol{\nabla}\psi\boldsymbol{\cdot}\boldsymbol{\nabla}\varphi\rangle$ with associated norm $\|\cdot\|_{1,2}$ given by $\|\psi\|_{1,2} = \langle|\boldsymbol{\nabla}\psi|^2\rangle^{1/2}$. We emphasize that in the case of a time-dependent fluid velocity field, it is necessary that $\psi \in \mathscr{H}$ satisfy $\langle\psi\rangle_{\mathcal{V}} = 0$, as required by the definition of $\mathscr{H}_{\mathcal{V}}^{1,2}$ in (A-15). Otherwise, $\|\cdot\|_{1,2} = \int_{\mathcal{T}\times\mathcal{V}} \mathrm{d}t\,\mathrm{d}\boldsymbol{x}\,|\boldsymbol{\nabla}\cdot|^2$ is not a norm, since a strictly positive function $\psi(t,\boldsymbol{x}) = \psi(t)$ on $\mathcal{T}\times\mathcal{V}$ satisfies

$\|\psi\|_{1,2} = 0$. In the case of a time-independent fluid velocity field $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$ we set $\mathscr{H} \equiv \mathscr{F} \equiv \mathscr{H}_{\mathcal{V}}^{1,2}$ and $\langle\cdot\rangle = \langle\cdot\rangle_{\mathcal{V}}$.

We now use properties of the Hilbert space $\mathscr{H}$ to obtain functional formulas for the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* defined in (10) and (11), involving the solution χ_j of the cell problem in (9) and a maximal skew-adjoint operator A on $\mathscr{F}$. We then derive from the cell problem a resolvent formula for χ_j involving A. The spectral theorem discussed in Appendix A then yields Stieltjes integral representations for S^* and A^*, which are established in Theorem 1 below.

Applying the linear operator $(-\Delta)^{-1}$ to both sides of the cell problem in equation (9) yields, with suitable notational changes,

$$(-\Delta)^{-1}u_j = (\varepsilon + A)\chi_j, \qquad A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla}). \tag{A-17}$$

We will discuss the key properties of the operators $(-\Delta)^{-1}$ and A in more detail below. Now write the functional $\langle u_j\chi_k\rangle$ in equation (10) as [79]

(A-18)
$$\langle u_j\chi_k\rangle = \langle[(-\Delta)(-\Delta)^{-1}u_j]\,\chi_k\rangle = \langle\boldsymbol{\nabla}(-\Delta)^{-1}u_j\cdot\boldsymbol{\nabla}\chi_k\rangle = \langle(-\Delta)^{-1}u_j, \chi_k\rangle_{1,2}.$$

This calculation will be justified below. Substituting the formula in (A-17) for $(-\Delta)^{-1}u_j$ into equation (A-18) yields equation (15), which provides functional formulas for the components S^*_{jk} and A^*_{jk}, $j,k = 1,\dots,d$, of S^* and A^*. Equation (A-17) leads to the resolvent formula shown in (16). From equations (15) and (16) we have the functional formulas for S^*_{jk} and A^*_{jk} shown in equation (17) involving the resolvent of the operator A. The following theorem establishes the Stieltjes integral representations in equation (18) for these functional formulas of S^*_{jk} and A^*_{jk}.

Theorem 1. *Assume $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}_{\mathcal{V}}^{0,2} \cap L^\infty(\mathcal{V}))$ for all $j = 1,\dots,d$. Then $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$ is a maximal (skew-adjoint) normal operator on the function space $\mathscr{F}$, hence $M = -\imath A$ is self-adjoint on $\mathscr{F}$. Let $Q(\lambda)$ be the resolution of the identity in one-to-one correspondence with M. Define the complex valued function $\mu_{jk}(\lambda) = \langle Q(\lambda)g_j, g_k\rangle_{1,2}$, $j,k = 1,\dots,d$, where $g_j = (-\Delta)^{-1}u_j$. Consider the positive measure μ_{kk} and the signed measures $Re\,\mu_{jk}$ and $Im\,\mu_{jk}$ associated with $\mu_{jk}(\lambda)$, introduced in equations (A-4) and (A-5). Then, for all $0 < \varepsilon < \infty$ the functional formulas for S^*_{jk} and A^*_{jk} in (17) have the Radon–Stieltjes integral representations shown in (18).*

Before we prove Theorem 1, we first provide in Appendix C.1.1 key properties of the linear operator $(-\Delta)^{-1}$ and justify the calculation in equation (A-18). Moreover, in Appendix C.1.2 we discuss key properties of the function u_j on the right side of the cell problem in (9). These properties of $(-\Delta)^{-1}$ and u_j will be used in the proof of Theorem 1, which is deferred to Appendix C.1.3.

C.1.1. Properties of the inverse Laplacian. Since the Laplacian Δ maps $\mathcal{V}$–periodic functions to *mean-zero* $\mathcal{V}$–periodic functions, in the present context, the domain of the operator $(-\Delta)^{-1}$ is mean-zero $\mathcal{V}$–periodic functions. The operator $(-\Delta)^{-1}$ is based on convolution with respect to the Green's function for the Laplacian, i.e., $(-\Delta)^{-1}f(\boldsymbol{x}) = \int_{\mathcal{V}} G(\boldsymbol{x}-\boldsymbol{y})\, f(\boldsymbol{y})\, \mathrm{d}\boldsymbol{y}$. The Green's function G is positive [95], $G > 0$, symmetric [33, 95] $G(\boldsymbol{x}-\boldsymbol{y}) = G(\boldsymbol{y}-\boldsymbol{x})$ and integrable [64, 65, 59],

$$\sup_{\boldsymbol{x}\in\mathcal{V}} \int_{\mathcal{V}} G(\boldsymbol{x}-\boldsymbol{y})\mathrm{d}\boldsymbol{y} \le C < \infty. \tag{A-19}$$

The Green's function G can be represented [64] in terms of the eigenvalues $\lambda_{\boldsymbol{k}}$ and orthonormal eigenfunctions $\phi_{\boldsymbol{k}}(\boldsymbol{x})$ of the operator $-\Delta$ with periodic boundary conditions on $\mathcal{V}$, $G(\boldsymbol{x}-\boldsymbol{y}) = \sum_{\boldsymbol{k}\in\mathbb{Z}^d\setminus\{0\}} \phi_{\boldsymbol{k}}(\boldsymbol{x})\, \phi_{\boldsymbol{k}}(\boldsymbol{y})/\lambda_{\boldsymbol{k}}$, where $\lambda_{\boldsymbol{k}} = |\boldsymbol{k}|^2$ and $\{\phi_{\boldsymbol{k}}(\boldsymbol{x})\} = \{\cos(\boldsymbol{k}\cdot\boldsymbol{x}), \sin(\boldsymbol{k}\cdot\boldsymbol{x})\}$ when $\mathcal{V} = [0, 2\pi]^d$.

By equation (A-19) and Young's inequality [33, 34], $(-\Delta)^{-1}$ is a bounded operator on $L^p(\mathcal{V})$ for $1 \le p \le \infty$: if $\psi \in L^p(\mathcal{V})$ then $(-\Delta)^{-1}\psi \in L^p(\mathcal{V})$ and

$$\|(-\Delta)^{-1}\psi\|_p \le C\|\psi\|_p, \quad 1 \le p \le \infty, \tag{A-20}$$

where $\|\cdot\|_p$ denotes the $L^p(\mathcal{V})$–norm and C is defined in (A-19). Since $\mathcal{V}$ is bounded, it has finite Lebesgue measure $|\mathcal{V}| < \infty$. Consequently, we have [34] $L^p(\mathcal{V}) \supset L^q(\mathcal{V})$ for all $0 < p < q \le \infty$ with $\|\psi\|_p \le \|\psi\|_q\, |\mathcal{V}|^{(1/p)-(1/q)}$.

Recall that $\mathscr{H}_{\mathcal{V}}^{0,2}$ is the closure of $C^2(\mathcal{V})$ in the norm $\|\cdot\|_{\mathcal{V}}$. For $f \in \mathscr{H}_{\mathcal{V}}^{0,2}$, the operator $(-\Delta)^{-1}$ satisfies $\langle(-\Delta)(-\Delta)^{-1}f, h\rangle_{\mathcal{V}} = \langle f, h\rangle_{\mathcal{V}}$ in the following weak sense [95, 88]. Let $\{f_n\} \subset C^2(\mathcal{V})$ be a sequence that is Cauchy in the norm $\|\cdot\|_{\mathcal{V}}$ with $\lim_{n\to\infty}\|f_n - f\|_{\mathcal{V}} = 0$. Then, for all $h \in \mathscr{H}_{\mathcal{V}}^{0,2}$ (see Theorem 1 in Section 4.2 of [63]),

$$\begin{aligned} \langle(-\Delta)(-\Delta)^{-1}f, h\rangle_{\mathcal{V}} &:= \lim_{n\to\infty}\left\langle \int_{\mathcal{V}} G(\boldsymbol{x}-\boldsymbol{y})(-\Delta_y)f_n(\boldsymbol{y})\mathrm{d}\boldsymbol{y}, h(\boldsymbol{x})\right\rangle_{\mathcal{V}} \\ &= \lim_{n\to\infty}\langle f_n, h\rangle_{\mathcal{V}} = \langle f, h\rangle_{\mathcal{V}}, \end{aligned} \tag{A-21}$$

by the continuity of the inner-product [34, 95] and since the boundary terms [63] $\int_{\partial\mathcal{V}}[f_n(\boldsymbol{y})\, \partial G(\boldsymbol{x}-\boldsymbol{y})/\partial\mathbf{n}_y - G(\boldsymbol{x}-\boldsymbol{y})\, \partial f_n(\boldsymbol{y})/\partial\mathbf{n}_y]\, \mathrm{d}S_y$ vanish by periodicity. Here, $\mathrm{d}S_y$ denotes the surface measure [34] on the boundary $\partial\mathcal{V}$

of $\mathcal{V}$ and $\partial/\partial\mathbf{n}_y$ is the outward normal derivative on $\partial\mathcal{V}$ [63]. Moreover, equation (A-21), integration by parts, Young's inequality in (A-20) for $p = 2$, and the Cauchy–Schwartz inequality, $|\langle f, g\rangle_{\mathcal{V}}| \le \|f\|_{\mathcal{V}}\,\|g\|_{\mathcal{V}}$, imply for $\psi \in \mathscr{H}^{0,2}_{\mathcal{V}}$

$$\|\boldsymbol{\nabla}(-\Delta)^{-1}\psi\|^2_{\mathcal{V}} = |\langle(-\Delta)^{-1}\psi, \psi\rangle_{\mathcal{V}}| \le C\|\psi\|^2_{\mathcal{V}}\,, \tag{A-22}$$

in the weak sense shown in equation (A-21).

We now justify the calculation in (A-18). If $u_j(t,\cdot), \chi_j(t,\cdot) \in \mathscr{H}^{0,2}_{\mathcal{V}}$ for almost all $t \in \mathcal{T}$, then equation (A-21) yields the first equality in (A-18). However, the second equality requires $\chi_j(t,\cdot) \in \mathscr{H}^{1,2}_{\mathcal{V}}$. We now establish $\mathscr{H}^{1,2}_{\mathcal{V}} \subset \mathscr{H}^{0,2}_{\mathcal{V}}$. This, in turn, justifies the calculation in equation (A-18) and, along with equation (A-22), also shows that the operator $(-\Delta)^{-1}$ maps functions in $\mathscr{H}^{0,2}_{\mathcal{V}}$ to $\mathscr{H}^{1,2}_{\mathcal{V}}$.

Recall that $\mathscr{H}^{0,2}_{\mathcal{V}}$ is the closure of $C^2(\mathcal{V})$ in the norm $\|\cdot\|_{\mathcal{V}}$, $\mathscr{H}^{1,2}_{\mathcal{V}}$ is the closure of $C^3(\mathcal{V})$ in the norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$, and $C^3(\mathcal{V}) \subset C^2(\mathcal{V})$. Let $\{f_n\}$ be an arbitrary sequence in $C^3(\mathcal{V})$ that is Cauchy in the norm $\|\boldsymbol{\nabla}\cdot\|_{\mathcal{V}}$. Then equation (A-21), integration by parts, Young's inequality in (A-20) for $p = 2$, and the Cauchy–Schwartz inequality $|\langle\boldsymbol{\xi}\boldsymbol{\cdot}\boldsymbol{\zeta}\rangle_{\mathcal{V}}| \le \|\boldsymbol{\xi}\|_{\mathcal{V}}\|\boldsymbol{\zeta}\|_{\mathcal{V}}$ imply

$$\|f_n - f_m\|_{\mathcal{V}} \le C\|\boldsymbol{\nabla}(f_n - f_m)\|_{\mathcal{V}}, \tag{A-23}$$

where the constant C is defined in (A-19). Consequently, every sequence $\{f_n\}$ in $C^3(\mathcal{V})$ that is Cauchy in the norm $\|\nabla\cdot\|_{\mathcal{V}}$ is also a sequence in $C^2(\mathcal{V})$ that is Cauchy in the norm $\|\cdot\|_{\mathcal{V}}$. This establishes $\mathscr{H}^{1,2}_{\mathcal{V}} \subset \mathscr{H}^{0,2}_{\mathcal{V}}$.

C.1.2. Properties of the forcing function in the cell problem. We now discuss key properties of the forcing function u_j on the right side of the cell problem in (9). Recall the discussion in Section 2.2 regarding the properties of the effective diffusivity tensor D^* when the fluid velocity field $\boldsymbol{u}$ has a large scale mean flow $\boldsymbol{u}_0$. Specifically, recall that the cell problem is analogous to equation (9), which holds for a mean-zero velocity field, $\boldsymbol{u}_0 \equiv 0$. Moreover, the function u_j on the right side of the cell problem is mean-zero, $\langle u_j\rangle = 0$, regardless of whether the strength of $\boldsymbol{u}_0$ is weak [56, 79] or comparable [79] to the periodic fluctuations, e.g. u_j is replaced by the jth component of the mean-zero vector field $\boldsymbol{u} - \boldsymbol{u}_0$.

Using the $\mathscr{H}$–inner-product to obtain Stieltjes integral representations for D^* requires the function u_j to be mean-zero in space alone, $\langle u_j\rangle_{\mathcal{V}} = 0$. In particular, in the proof of Theorem 1 below we show it is required that $(-\Delta)^{-1}u_j \in \mathscr{F}$, where $\mathscr{F} = \tilde{\mathscr{A}}_{\mathcal{T}} \otimes \mathscr{H}^{1,2}_{\mathcal{V}}$. Recall for $\psi \in \mathscr{H}^{1,2}_{\mathcal{V}}$ it is required that $\langle\psi\rangle_{\mathcal{V}} = 0$. We therefore require $\langle(-\Delta)^{-1}u_j(t,\cdot)\rangle_{\mathcal{V}} = 0$ for almost all

$t \in \mathcal{T}$. Since the domain of the operator $(-\Delta)^{-1}$ is mean-zero $\mathcal{V}$–periodic functions, we require $\langle u_j(t,\cdot)\rangle_{\mathcal{V}} = 0$ for almost all $t \in \mathcal{T}$.

Conversely, $\psi \in \mathscr{H}_{\mathcal{V}}$ and $\langle \psi \rangle_{\mathcal{V}} = 0$ imply $\langle (-\Delta)^{-1}\psi\rangle_{\mathcal{V}} = 0$. More specifically, since the eigenfunctions $\phi_{\boldsymbol{k}}$ of $-\Delta$ are mean-zero, $\mathcal{V}$–periodic, and $\phi_{\boldsymbol{k}} \in C^\infty(\mathcal{V})$, $-\Delta\phi_{\boldsymbol{k}} = |\boldsymbol{k}|^2\phi_{\boldsymbol{k}}$ implies $(-\Delta)^{-1}\phi_{\boldsymbol{k}} = |\boldsymbol{k}|^{-2}\phi_{\boldsymbol{k}}$, hence $\langle(-\Delta)^{-1}\phi_{\boldsymbol{k}}\rangle_{\mathcal{V}} = 0$ (see Theorem 1 in Section 4.2 of [63]). Since $\{\phi_{\boldsymbol{k}} \,|\, \boldsymbol{k} \in \mathbb{Z}^d\}$ is a complete orthonormal basis for $\mathscr{H}_{\mathcal{V}}$ [34], $\psi \in \mathscr{H}_{\mathcal{V}}$ and $\langle\psi\rangle_{\mathcal{V}} = 0$ imply the sequence of functions ψ_n defined by $\psi_n = \sum_{|\boldsymbol{k}|\le n}\langle \psi, \phi_{\boldsymbol{k}}\rangle_{\mathcal{V}}\,\phi_{\boldsymbol{k}}$ converges to ψ in norm, $\lim_{n\to\infty}\|\psi-\psi_n\|_{\mathcal{V}} \to 0$. Moreover $\langle(-\Delta)^{-1}\psi_n\rangle_{\mathcal{V}} = 0$ for all $n \in \mathbb{N}$. Therefore, equation (A-20), $L^1(\mathcal{V}) \supset L^2(\mathcal{V})$, and the triangle inequality imply $|\langle(-\Delta)^{-1}\psi\rangle_{\mathcal{V}}| = |\langle(-\Delta)^{-1}(\psi-\psi_n)\rangle_{\mathcal{V}}| \le C\|\psi-\psi_n\|_1 \to 0$ as $n\to\infty$. This implies $\langle(-\Delta)^{-1}\psi\rangle_{\mathcal{V}} = 0$ for all mean-zero $\psi \in \mathscr{H}_{\mathcal{V}}$.

The result of Lemma 2 below is used in the proof of Theorem 1. There, we assume $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes L^\infty(\mathcal{V})$. This property, $\langle u_j(t,\cdot)\rangle_{\mathcal{V}} = 0$ for almost all $t \in \mathcal{T}$, $\tilde{\mathscr{A}}_{\mathcal{T}} \otimes L^\infty(\mathcal{V}) \subset L^2(\mathcal{T}\times\mathcal{V})$, and the Fubini-Tonelli theorem [34] imply that $\langle u_j\rangle = 0$. We stress it is *not* necessary that $\langle u_j(\cdot,\boldsymbol{x})\rangle_{\mathcal{T}} = 0$ for almost all $\boldsymbol{x}\in\mathcal{V}$. The fluid velocity field $\boldsymbol{u}$ in equation (1) is such an example, satisfying $\langle\boldsymbol{u}\rangle_{\mathcal{V}} = 0$ and $\langle\boldsymbol{u}\rangle = 0$ but $\langle\boldsymbol{u}\rangle_{\mathcal{T}} = (\cos y, \cos x) \not\equiv 0$. The condition $\langle u_j\rangle_{\mathcal{V}} = 0$ rules out functions of the form $u_j(t,\boldsymbol{x}) = u_j(t)$ or even $u_j(t,\boldsymbol{x}) = f(t,\boldsymbol{x}) + h(t)$ with $\langle f\rangle_{\mathcal{V}} = 0$, though functions of the form $u_j(t,\boldsymbol{x}) = h(t)\,f(t,\boldsymbol{x})$ with $\langle f\rangle_{\mathcal{V}} = 0$ are permitted.

In summary, the properties we require the function u_j on the right side of the cell problem in equation (9) to have are: u_j is $\mathcal{T}\times\mathcal{V}$–periodic, $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V}))$, and $\langle u_j(t,\cdot)\rangle_{\mathcal{V}} = 0$ for almost all $t\in\mathcal{T}$, (which implies that $\langle(-\Delta)^{-1}u_j(t,\cdot)\rangle_{\mathcal{V}} = \langle(-\Delta)^{-1}u_j(t,\cdot)\rangle = 0$ and $\langle u_j\rangle = 0$). Note, since every $f \in C^2(\mathcal{V})$ is also a member of $\mathscr{H}^{0,2}_{\mathcal{V}}$ and $L^\infty(\mathcal{V})$, the intersection $\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V})$ is non-empty.

The following lemma identifies a sufficient condition for the fluid velocity field $\boldsymbol{u}$ that ensures the operator $(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})$ is bounded on the Hilbert space $\mathscr{H}$. This will be used in the proof of Theorem 1 below.

Lemma 2. *Assume the components u_j, $j = 1,\dots,d$, of the fluid velocity field $\boldsymbol{u}$ satisfy $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V}))$. Then the operator $(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})$ is bounded on $\mathscr{H}$. Moreover, its operator norm $\|(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})\|_{1,2}$ has the following upper bound*

$$\|(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})\|_{1,2} \le \sqrt{C}\,\|\,|\boldsymbol{u}|\,\|_\infty\,, \tag{A-24}$$

where $\|\cdot\|_\infty$ the $L^\infty(\mathcal{T}\times\mathcal{V})$–norm and the constant C is defined in equation (A-19) and satisfies $0 < C < \infty$.

Proof of Lemma 2. The spectral theorem used in the proof of Theorem 1 below requires that the operator $\boldsymbol{u}\cdot\boldsymbol{\nabla}$ acts on the Hilbert space $\mathscr{H}$ and $(-\Delta)^{-1}u_j \in \mathscr{F}$, where $\mathscr{H} = \mathscr{H}_{\mathcal{T}} \otimes \mathscr{H}^{1,2}_{\mathcal{V}}$ and $\mathscr{F} = \tilde{\mathscr{A}}_{\mathcal{T}} \otimes \mathscr{H}^{1,2}_{\mathcal{V}}$. Consequently, based on the discussion in the paragraph following equation (A-22), we require that $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes \mathscr{H}^{0,2}_{\mathcal{V}}$. Moreover, writing $\boldsymbol{u}\cdot\boldsymbol{\nabla} f$ requires $f \in \mathscr{H}$. Recall, that $\mathscr{H}^{1,2}_{\mathcal{V}} \subset \mathscr{H}^{0,2}_{\mathcal{V}} \subset L^2(\mathcal{V})$, $\mathscr{H} \subset L^2(\mathcal{T}) \otimes L^2(\mathcal{V})$, and $L^2(\mathcal{V}) \supseteq L^r(\mathcal{V})$ for all $2 \le r \le \infty$, and similarly for $L^2(\mathcal{T})$ and $L^2(\mathcal{T}\times\mathcal{V})$.

We now address the following question: when $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^r(\mathcal{V}))$ for some $2 \le r \le \infty$ and $f \in \mathscr{H}$, for what value of p, $1 \le p \le \infty$, is $\boldsymbol{u}\cdot\boldsymbol{\nabla} f \in L^p(\mathcal{T}\times\mathcal{V})$? Denote by $\|\cdot\|_p$ the $L^p(\mathcal{T}\times\mathcal{V})$–norm. The Cauchy–Schwartz inequality, $|\boldsymbol{\xi}\cdot\boldsymbol{\zeta}| \le |\boldsymbol{\xi}|\,|\boldsymbol{\zeta}|$ and Hölder's inequality [34], $\|f\,h\|_1 \le \|f\|_{p_1}\,\|h\|_{q_1}$, with conjugate exponents satisfying $(1/p_1) + (1/q_1) = 1$ and $1 \le p_1 \le 2 \le q_1 \le \infty$, yield

$$\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_p^p \le \|\,|\boldsymbol{u}|^{p\,p_1}\|_1^{1/p_1}\,\||\boldsymbol{\nabla} f|^{p\,q_1}\|_1^{1/q_1}. \tag{A-25}$$

We require $p\,q_1 = 2$ and write $r = p\,p_1$. This and $(1/p_1) + (1/q_1) = 1$ yield

$$\frac{1}{p} = \frac{1}{r} + \frac{1}{2}, \qquad 2 \le r \le \infty, \quad 1 \le p \le 2. \tag{A-26}$$

We now use equations (A-25) and (A-26) with $r = \infty$ to establish the bound in equation (A-24).

Consider equation (A-21) with the functions f and h both given by $\boldsymbol{u}\cdot\boldsymbol{\nabla} f$ with $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ and $u_j(t,\cdot) \in \mathscr{H}^{0,2}_{\mathcal{V}}$ for almost all $t \in \mathcal{T}$. This ensures that the Cauchy sequence $\{\psi_n\}$, say, associated with the function $\boldsymbol{u}\cdot\boldsymbol{\nabla} f$ has members satisfying $\psi_n(t,\cdot) \in C^2(\mathcal{V})$ for almost all $t \in \mathcal{T}$. Consequently, integration by parts, Young's inequality in (A-20), and Hölder's inequality $|\langle f\,h\rangle| \le \|f\|_{p_2}\,\|h\|_{q_2}$ with $(1/p_2) + (1/q_2) = 1$ and $1 \le p_2 \le 2 \le q_2 \le \infty$, yield

$$\begin{aligned} \|(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})f\|_{1,2}^2 &= |\langle[(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla} f)]\,(\boldsymbol{u}\cdot\boldsymbol{\nabla} f)\rangle| \\ &\le \|(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla} f)\|_{p_2}\,\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{q_2} \\ &\le C\,\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{p_2}\,\|\boldsymbol{u}\cdot\boldsymbol{\nabla} f\|_{q_2}. \end{aligned} \tag{A-27}$$

Setting $p_2 = q_2 = 2$, equations (A-25) and (A-26) with $r = \infty$ ($p_1 = \infty$ and $q_1 = 1$) establishes the bound in (A-24). This completes the proof of Lemma 2. $\square$

C.1.3. Proof of Theorem 1. We first establish that the operator $M = -\imath A$ with domain $\mathscr{F}$ is self-adjoint, where $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$. We have already established in Appendix B that the operator $-\imath\partial_t$ with domain $\tilde{\mathscr{A}}_{\mathcal{T}}$ is self-adjoint [97]. A bounded linear symmetric operator is self-adjoint on a Hilbert space if and only if its domain is the Hilbert space itself (see Theorem 2.24 in [97]). By Young's inequality in (A-20), the linear operator $(-\Delta)^{-1}$ is bounded on the space of functions $\mathscr{H}_{\mathcal{V}}$ with mean-zero, $\mathscr{H}_{\mathcal{V}}\backslash\mathbb{C}$. It is also symmetric on $\mathscr{H}_{\mathcal{V}}\backslash\mathbb{C}$ [95, 33]. Consequently, the operator $(-\Delta)^{-1}$ with domain $\mathscr{H}_{\mathcal{V}}\backslash\mathbb{C}$ is self-adjoint. It is also self-adjoint on $\mathscr{H}^{1,2}_{\mathcal{V}}$. Indeed, recalling that $\mathcal{V} = [0, L]^d$, the calculation in equation (A-22) and the Poincaré inequality [63], $\|f\|_{\mathcal{V}} \le 2L\|\boldsymbol{\nabla} f\|_{\mathcal{V}}$, show that the operator $(-\Delta)^{-1}$ is bounded on $\mathscr{H}^{1,2}_{\mathcal{V}}$ with operator norm bounded by the quantity $2L\sqrt{C}$. It is also symmetric on the Hilbert space $\mathscr{H}^{1,2}_{\mathcal{V}}$, as the following calculation shows, which establishes that the operator $(-\Delta)^{-1}$ with domain $\mathscr{H}^{1,2}_{\mathcal{V}}$ is self-adjoint. Similar to (A-22) for $f, h \in \mathscr{H}^{1,2}_{\mathcal{V}}$ we have

(A-28)
$$\langle\boldsymbol{\nabla}(-\Delta)^{-1}f\cdot\boldsymbol{\nabla}h\rangle_{\mathcal{V}} = \langle f, h\rangle_{\mathcal{V}} = \langle f, (-\Delta)(-\Delta)^{-1}h\rangle_{\mathcal{V}} = \langle\boldsymbol{\nabla}f\cdot\boldsymbol{\nabla}(-\Delta)^{-1}h\rangle_{\mathcal{V}}.$$

By equation (A-20), the operators $-\imath\partial_t$ and $(-\Delta)^{-1}$ commute on $\mathscr{F} = \tilde{\mathscr{A}}_{\mathcal{T}} \otimes \mathscr{H}^{1,2}_{\mathcal{V}}$ (see Theorem 2.27 in [34]). By equation (A-22), the range of the operator $(-\Delta)^{-1}$ with domain $\mathscr{H}^{0,2}_{\mathcal{V}}$ is contained in $\mathscr{H}^{1,2}_{\mathcal{V}}$. Consequently, the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F} \cap \mathscr{H} = \mathscr{F}$ [97] is self-adjoint.

In Lemma 2 we established that the linear operator $(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is bounded when $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V}))$. We now show that this condition on u_j implies that the operator is also antisymmetric on $\mathscr{H}$ which, in turn, establishes that the symmetric operator $-\imath(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is self-adjoint. The antisymmetry of $(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]$ on $\mathscr{H}$ depends on the incompressibility, $\boldsymbol{\nabla}\cdot\boldsymbol{u} = 0$ (weakly), of the fluid velocity field [12]. For $f, h \in \mathscr{H}$ we have, in a weak sense,

$$\begin{aligned}
\text{(A-29)} \qquad \langle(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})f, h\rangle_{1,2} &= \langle[\boldsymbol{\nabla}((-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla}f)]\cdot\boldsymbol{\nabla}h\rangle \\
&= \langle[(\boldsymbol{u}\cdot\boldsymbol{\nabla}f)]\,, h\rangle \\
&= \langle[\boldsymbol{\nabla}\cdot(\boldsymbol{u}f)]\,, h\rangle \\
&= -\langle f\,, [(\boldsymbol{u}\cdot\boldsymbol{\nabla})h]\rangle \\
&= -\langle f\,, [(-\Delta)(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})h]\rangle \\
&= -\langle\boldsymbol{\nabla}f\cdot[\boldsymbol{\nabla}(-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})h]\rangle \\
&= -\langle f, (-\Delta)^{-1}(\boldsymbol{u}\cdot\boldsymbol{\nabla})h\rangle_{1,2}.
\end{aligned}$$

This establishes that the bounded linear operator $-\imath(-\Delta)^{-1}(\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})$ is symmetric on $\mathscr{H}$, hence self-adjoint on $\mathscr{H}$.

We now summarize our findings. We have established that the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F}$ is self-adjoint and the operator $-\imath(-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is self-adjoint when the components u_j, $j=1,\ldots,d$, of $\boldsymbol{u}$ satisfy $u_j\in\tilde{\mathscr{A}_{\mathcal{T}}}\otimes(\mathscr{H}_{\mathcal{V}}^{0,2}\cap L^\infty(\mathcal{V}))$. Consequently, the difference of these two operators $M=-\imath A$, $A=(-\Delta)^{-1}(\partial_t-\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla})$, with domain $D(M)=\mathscr{F}\cap\mathscr{H}=\mathscr{F}$ [97] is self-adjoint when $u_j\in\tilde{\mathscr{A}_{\mathcal{T}}}\otimes(\mathscr{H}_{\mathcal{V}}^{0,2}\cap L^\infty(\mathcal{V}))$. Thus $A=\imath M$ is a maximal (skew-adjoint) normal operator on $\mathscr{F}$ [97].

The complex-valued functions involved in the functional formulas for S^*_{jk} and A^*_{jk} in (17) are $F(\lambda)=(\varepsilon+\imath\lambda)^{-1}$ and $G(\lambda)=\imath\lambda(\varepsilon+\imath\lambda)^{-1}$. For all $0<\varepsilon<\infty$, we have $|F(\lambda)|^2=(\varepsilon^2+\lambda^2)^{-1}\le\varepsilon^{-2}<\infty$ and $|G(\lambda)|^2=\lambda^2(\varepsilon^2+\lambda^2)^{-1}\le 1$. Since μ_{kk} is a finite measure for all $k=1,\ldots,d$, as shown in equation (A-7), we therefore have that $f\in\mathscr{D}(F)$ and $f\in\mathscr{D}(G)$ for all $f\in D(M)$ when $0<\varepsilon<\infty$.

We now establish that the function $g_j=(-\Delta)^{-1}u_j$ satisfies $g_j\in D(M)$. By assumption, u_j is $\mathcal{T}\times\mathcal{V}$–periodic and satisfies $\langle u_j\rangle_{\mathcal{V}}=0$. From the discussion in Appendix C.1.2 we have $\langle g_j\rangle_{\mathcal{V}}=0$. By assumption, $u_j\in\tilde{\mathscr{A}_{\mathcal{T}}}\otimes(\mathscr{H}_{\mathcal{V}}^{0,2}\cap L^\infty(\mathcal{V}))$. By equation (A-22) the operator $(-\Delta)^{-1}$ maps $\mathscr{H}_{\mathcal{V}}^{0,2}$ to $\mathscr{H}_{\mathcal{V}}^{1,2}$. Consequently, we have $g_j\in\mathscr{F}$, where $\mathscr{F}=\tilde{\mathscr{A}_{\mathcal{T}}}\otimes\mathscr{H}_{\mathcal{V}}^{1,2}$. Since $\mathscr{F}\subseteq D(M)$ we have $g_j\in D(M)$.

The conditions of the spectral theorem are thus satisfied. Consequently, the integral representations in equation (A-6) hold for the functions $F(\lambda)$ and $G(\lambda)$ defined above, involving the complex measure μ_{jk}. The discussion leading to equation (A-9) then establishes the integral representations for S^*_{jk} and A^*_{jk} shown in equation (18).

It is worth noting that from equations (A-7) and (A-22), the mass μ^0_{jk} of the measure μ_{jk} is given by $\mu^0_{jk}=\langle g_j,g_k\rangle_{1,2}=\langle(-\Delta)^{-1}u_j,u_k\rangle$. Since $u_j\in\tilde{\mathscr{A}_{\mathcal{T}}}\otimes\mathscr{H}_{\mathcal{V}}$ and $(-\Delta)^{-1}$ is a self-adjoint operator on $\mathscr{H}_{\mathcal{V}}$, hence on $\tilde{\mathscr{A}_{\mathcal{T}}}\otimes\mathscr{H}_{\mathcal{V}}$, the spectral theorem demonstrates

$$\mu^0_{jk}=\langle(-\Delta)^{-1}u_j,u_k\rangle=\int\lambda\,\mathrm{d}\langle\tilde{Q}(\lambda)u_j,u_k\rangle. \tag{A-30}$$

In other words, the mass μ^0_{jk} of the measure μ_{jk} is the first moment of the spectral measure $\mathrm{d}\langle\tilde{Q}(\lambda)u_j,u_k\rangle$ for the operator $(-\Delta)^{-1}$, where $\tilde{Q}(\lambda)$ is the resolution of the identity in one-to-one correspondence with the self-adjoint operator $(-\Delta)^{-1}$. This completes the proof of Theorem 1. □

C.2. Curl-free vector fields and effective diffusivity

In this section we consider an alternate formulation of the effective parameter problem for advection-diffusion that was first proposed [4, 5] for *time-independent* flows. In particular, we provide a rigorous mathematical framework which generalizes this formulation to include space-time periodic fluid velocity fields that are allowed to have chaotic dynamics. This approach provides analogous formulas to those shown in equations (15)–(18) involving the *curl-free* vector field $\boldsymbol{\nabla}\chi_j$ shown in equation (9), with suitable notational changes, and a maximal (skew-adjoint) normal operator $\mathbf{A}$ acting on a Hilbert space of vector-valued functions.

Towards this goal, recall the Hilbert spaces $\mathscr{H}_\mathcal{T}$ and $\mathscr{H}_\mathcal{V}$ of scalar functions given in equation (A-14) and the function space $\tilde{\mathscr{A}}_\mathcal{T}$ given in (A-13). Now define their d-dimensional analogues over the complex field $\mathbb{C}$,

$$\mathcal{H}_\mathcal{T}=\otimes_{j=1}^{d}\mathscr{H}_\mathcal{T},\qquad \mathcal{H}_\mathcal{V}=\otimes_{j=1}^{d}\mathscr{H}_\mathcal{V},\qquad \tilde{\mathcal{A}}_\mathcal{T}=\otimes_{j=1}^{d}\tilde{\mathscr{A}}_\mathcal{T}. \tag{A-31}$$

The Hilbert space $\mathcal{H}_\mathcal{V}$ can be decomposed into mutually orthogonal subspaces of (weakly) curl-free $\mathcal{H}_\times$, divergence-free $\mathcal{H}_\bullet$, and constant $\mathcal{H}_0$ vector fields, $\mathcal{H}_\mathcal{V}=\mathcal{H}_\times\oplus\mathcal{H}_\bullet\oplus\mathcal{H}_0$ [29, 68]. The orthogonal projectors associated with this decomposition are given by $\boldsymbol{\Gamma}_\times=-\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}$, $\boldsymbol{\Gamma}_\bullet=\boldsymbol{\nabla}\times(-\boldsymbol{\Delta})^{-1}\boldsymbol{\nabla}\times$, and $\boldsymbol{\Gamma}_0=\langle\cdot\rangle_\mathcal{V}$, respectively, satisfying $\mathsf{I}=\boldsymbol{\Gamma}_\times+\boldsymbol{\Gamma}_\bullet+\boldsymbol{\Gamma}_0$ [29, 74, 68]. Here, $\boldsymbol{\Delta}=\mathrm{diag}(\Delta,\dots,\Delta)$ is the vector Laplacian with inverse $\boldsymbol{\Delta}^{-1}=\mathrm{diag}(\Delta^{-1},\dots,\Delta^{-1})$, $\langle\cdot\rangle_\mathcal{V}$ denotes spatial averaging over $\mathcal{V}$, and I is the identity operator on $\mathcal{H}_\mathcal{V}$.

Using the *curl-free* vector field $\boldsymbol{\nabla}\chi_j$ in the cell problem in equation (9), we define the Hilbert space $\mathcal{H}_\times$ as

$$\mathcal{H}_\times=\{\boldsymbol{\psi}\in\mathcal{H}_\mathcal{V}\mid\boldsymbol{\Gamma}\boldsymbol{\psi}=\boldsymbol{\psi}\text{ weakly, }\langle\boldsymbol{\psi}\rangle_\mathcal{V}=0\},\quad \boldsymbol{\Gamma}=-\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}\,. \tag{A-32}$$

Here, we have denoted $\boldsymbol{\Gamma}_\times$ by $\boldsymbol{\Gamma}$ for notational simplicity and will continue to do so. Moreover, the requirement $\langle\boldsymbol{\psi}\rangle_\mathcal{V}=0$ is due to $\mathcal{H}_\times$ and $\mathcal{H}_0$ being orthogonal spaces. Analogous to equation (A-16), we define the Hilbert space $\mathcal{H}$ and its dense subset $\mathcal{F}$,

$$\mathcal{H}=\mathcal{H}_\mathcal{T}\otimes\mathcal{H}_\times,\qquad \mathcal{F}=\tilde{\mathcal{A}}_\mathcal{T}\otimes\mathcal{H}_\times. \tag{A-33}$$

We emphasize again that due to the presence of $\tilde{\mathscr{A}}_\mathcal{T}$ in the definition of $\mathcal{F}$, it is an everywhere dense subset of the Hilbert space $\mathcal{H}$, and not a Hilbert space itself. Recall that $\langle\cdot\rangle$ denotes space-time averaging over $\mathcal{T}\times\mathcal{V}$. Denote by $\langle\cdot,\cdot\rangle_\times$ the sesquilinear inner-product associated with the Hilbert space $\mathcal{H}$, defined as $\langle\boldsymbol{\psi},\boldsymbol{\varphi}\rangle_\times = \langle\boldsymbol{\psi}\cdot\boldsymbol{\varphi}\rangle$, with $\langle\boldsymbol{\psi},\boldsymbol{\varphi}\rangle_\times = \overline{\langle\boldsymbol{\varphi},\boldsymbol{\psi}\rangle}_\times$. Here, $\boldsymbol{\psi}\cdot\boldsymbol{\varphi} = \boldsymbol{\psi}^T\overline{\boldsymbol{\varphi}}$, transposition of the vector $\boldsymbol{\psi}$ is denoted $\boldsymbol{\psi}^T$, and $\overline{\boldsymbol{\varphi}}$ denotes component-wise complex conjugation, with $\boldsymbol{\psi}\cdot\boldsymbol{\psi} = |\boldsymbol{\psi}|^2$. The norm $\|\cdot\|_\times$ induced by this inner-product is given by $\|\boldsymbol{\psi}\|_\times = \langle\boldsymbol{\psi},\boldsymbol{\psi}\rangle_\times^{1/2}$. In the case of a steady fluid velocity field $\boldsymbol{u}=\boldsymbol{u}(\boldsymbol{x})$, we set $\mathcal{H}\equiv\mathcal{F}\equiv\mathcal{H}_\times$ and $\langle\cdot\rangle = \langle\cdot\rangle_\mathcal{V}$.

Recall that the Sobolev space $\mathscr{H}_\mathcal{V}^{1,2}$ in equation (A-15) is the closure in the norm $\|\boldsymbol{\nabla}\cdot\|_\mathcal{V}$ of the space $C^3(\mathcal{V})$ of all three times continuously differentiable functions in $\mathscr{H}_\mathcal{V}$ which are also mean-zero and $\mathcal{V}$–periodic [12]. If $f\in\mathscr{H}_\mathcal{V}^{1,2}$, equation (A-21) shows that $\boldsymbol{\nabla}f$ is curl free, $\boldsymbol{\nabla}f\in\mathcal{H}_\times$, in the following weak sense. Let $\{f_n\}$ be a sequence of mean-zero $\mathcal{V}$–periodic functions with $f_n\in C^3(\mathcal{V})$ that is Cauchy in the norm $\langle|\boldsymbol{\nabla}\cdot|^2\rangle_\mathcal{V}$ and converges to f in $L^2(\mathcal{V})$. Then, for all $\boldsymbol{\psi}\in\mathcal{H}_\times$ we have

(A-34)
$$\langle\boldsymbol{\Gamma}\boldsymbol{\nabla}f\cdot\boldsymbol{\psi}\rangle_\mathcal{V} := \lim_{n\to\infty}\langle\boldsymbol{\nabla}(-\Delta)^{-1}(-\Delta)f_n\cdot\boldsymbol{\psi}\rangle_\mathcal{V} = \lim_{n\to\infty}\langle\boldsymbol{\nabla}f_n\cdot\boldsymbol{\psi}\rangle_\mathcal{V} = \langle\boldsymbol{\nabla}f\cdot\boldsymbol{\psi}\rangle_\mathcal{V}.$$

Consequently, since the differential operator $\boldsymbol{\nabla}$ maps $\mathscr{H}_\mathcal{V}^{1,2}$ to $\mathcal{H}_\mathcal{V}\backslash\mathbb{C}^d$ we have $\{\boldsymbol{\nabla}f\in\mathcal{H}_\mathcal{V}\backslash\mathbb{C}^d \,|\, f\in\mathscr{H}_\mathcal{V}^{1,2}\}\subseteq\mathcal{H}_\times$. It is therefore clear that on the function space $\{\boldsymbol{\nabla}f\in\mathcal{H}_\mathcal{V}\backslash\mathbb{C}^d \,|\, f\in\mathscr{H}_\mathcal{V}^{1,2}\}$ the operator $\boldsymbol{\Gamma}$ is a projection, hence bounded by unity in operator norm and trivially symmetric (since it acts as the identity operator on $\mathcal{H}_\times$). This establishes a direct link between the Hilbert spaces $\mathscr{H}_\mathcal{V}^{1,2}$ and $\mathcal{H}_\times$. The following lemma shows that these Hilbert spaces are in one-to-one isometric correspondence. This establishes that $\mathcal{H}_\times\equiv\{\boldsymbol{\nabla}f\in\mathcal{H}_\mathcal{V}\backslash\mathbb{C}^d \,|\, f\in\mathscr{H}_\mathcal{V}^{1,2}\}$ which, in turn, establishes that the linear symmetric bounded operator $\boldsymbol{\Gamma}$ with domain $\mathcal{H}_\times$ is self-adjoint.

Lemma 3. *The Hilbert spaces $\mathscr{H}_\mathcal{V}^{1,2}$ and $\mathcal{H}_\times$ are in one-to-one isometric correspondence, which we denote by $\mathscr{H}_\mathcal{V}^{1,2}\sim\mathcal{H}_\times$. More specifically,* temporarily *denote the inner-product induced norm of the Hilbert space $\mathscr{H}_\mathcal{V}^{1,2}$ by $\|f\|_{1,2} = \langle\boldsymbol{\nabla}f\cdot\boldsymbol{\nabla}f\rangle_\mathcal{V}^{1/2}$ and the inner-product induced norm of the Hilbert space $\mathcal{H}_\times$ by $\|\boldsymbol{\psi}\|_\times = \langle\boldsymbol{\psi}\cdot\boldsymbol{\psi}\rangle_\mathcal{V}^{1/2}$. Then, for every $f\in\mathscr{H}_\mathcal{V}^{1,2}$ we have $\boldsymbol{\nabla}f\in\mathcal{H}_\times$ and $\|\boldsymbol{\nabla}f\|_\times = \|f\|_{1,2}$. Conversely, for every $\boldsymbol{\psi}\in\mathcal{H}_\times$ there exists unique $f\in\mathscr{H}_\mathcal{V}^{1,2}$ (up to equivalence class) such that $\boldsymbol{\psi}=\boldsymbol{\nabla}f$ and $\|f\|_{1,2} = \|\boldsymbol{\psi}\|_\times$.*

Proof of Lemma 3. The discussion involving equation (A-34) shows that if $f\in\mathscr{H}_\mathcal{V}^{1,2}$, then the vector field $\boldsymbol{\nabla}f\in\mathcal{H}_\mathcal{V}\backslash\mathbb{C}^d$ satisfies $\boldsymbol{\Gamma}\boldsymbol{\nabla}f=\boldsymbol{\nabla}f$ weakly so that $\boldsymbol{\nabla}f\in\mathcal{H}_\times$. Moreover, $\|\boldsymbol{\nabla}f\|_\times^2 = \langle\boldsymbol{\nabla}f\cdot\boldsymbol{\nabla}f\rangle_\mathcal{V} = \|f\|_{1,2}^2 < \infty$. Consequently, for every $f\in\mathscr{H}_\mathcal{V}^{1,2}$ we have $\boldsymbol{\nabla}f\in\mathcal{H}_\times$ and $\|\boldsymbol{\nabla}f\|_\times^2 = \|f\|_{1,2}^2$.

Conversely, $\boldsymbol{\psi} \in \mathcal{H}_\times$ implies $\boldsymbol{\psi} = \boldsymbol{\Gamma}\boldsymbol{\psi} = \boldsymbol{\nabla} f$ weakly, where we have defined the scalar-valued function $f = \Delta^{-1}\boldsymbol{\nabla}\cdot\boldsymbol{\psi}$. Since $\boldsymbol{\psi} = \boldsymbol{\nabla} f$, the $\mathscr{H}^{1,2}_{\mathcal{V}}$ norm of f satisfies $\|f\|^2_{1,2} = \langle\boldsymbol{\psi}\cdot\boldsymbol{\psi}\rangle_{\mathcal{V}} = \|\boldsymbol{\psi}\|^2_\times < \infty$ so that $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$. Moreover, f is uniquely determined by $\boldsymbol{\psi}$ (up to a zero Lebesgue measure equivalence class), since if $f_1 = \Delta^{-1}\boldsymbol{\nabla}\cdot\boldsymbol{\psi}$ and $f_2 = \Delta^{-1}\boldsymbol{\nabla}\cdot\boldsymbol{\psi}$ then $\boldsymbol{\Gamma}\boldsymbol{\psi} = \boldsymbol{\psi}$ implies that $\|f_1 - f_2\|_{1,2} = \|\boldsymbol{\psi} - \boldsymbol{\psi}\|_\times = 0$. Consequently, for every $\boldsymbol{\psi} \in \mathcal{H}_\times$ there exists unique $f \in \mathscr{H}^{1,2}_{\mathcal{V}}$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\|f\|_{1,2} = \|\boldsymbol{\psi}\|_\times$. In summary, the Hilbert spaces $\mathscr{H}^{1,2}_{\mathcal{V}}$ and $\mathcal{H}_\times$ are in one-to-one isometric correspondence, which we denote by $\mathscr{H}^{1,2}_{\mathcal{V}} \sim \mathcal{H}_\times$. This concludes our proof of Lemma 3. $\square$

Since the fluid velocity field $\boldsymbol{u}$ is incompressible, $\boldsymbol{\nabla}\cdot\boldsymbol{u} = 0$ (weakly), there is a real skew-symmetric matrix $\mathsf{H}(t, \boldsymbol{x})$ satisfying [4, 5]

$$\boldsymbol{u} = \boldsymbol{\nabla}\cdot\mathsf{H}, \qquad \mathsf{H}^T = -\mathsf{H}. \tag{A-35}$$

Note that $\boldsymbol{\nabla}\cdot[\mathsf{H}\boldsymbol{\nabla}\varphi] = [\boldsymbol{\nabla}\cdot\mathsf{H}]\cdot\boldsymbol{\nabla}\varphi + \mathsf{H} : \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi$. Due to the anti-symmetry of the matrix H and the symmetry of the Hessian operator $\boldsymbol{\nabla}\boldsymbol{\nabla}$ when acting on a sufficiently smooth space of functions, we have $\mathsf{H} : \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi = 0$ for all such smooth functions φ, yielding

$$\boldsymbol{\nabla}\cdot[\mathsf{H}\boldsymbol{\nabla}\varphi] = [\boldsymbol{\nabla}\cdot\mathsf{H}]\cdot\boldsymbol{\nabla}\varphi = \boldsymbol{u}\cdot\boldsymbol{\nabla}\varphi. \tag{A-36}$$

Using this identity and the representation of the velocity field $\boldsymbol{u}$ in (A-35), the advection-diffusion in equation (2) can be written as a diffusion equation [29, 74],

$$\partial_t\phi = \boldsymbol{\nabla}\cdot\mathsf{D}\boldsymbol{\nabla}\phi, \quad \phi(0, \boldsymbol{x}) = \phi_0(\boldsymbol{x}), \qquad \mathsf{D} = \varepsilon\mathsf{I} + \mathsf{H}, \tag{A-37}$$

where $\mathsf{D}(t, \boldsymbol{x}) = \varepsilon\mathsf{I} + \mathsf{H}(t, \boldsymbol{x})$ can be viewed as a local diffusivity tensor with coefficients

$$\mathsf{D}_{jk} = \varepsilon\delta_{jk} + \mathsf{H}_{jk}, \quad j, k = 1, \ldots, d. \tag{A-38}$$

The cell problem in (9) can also be written as the following diffusion equation [29, 74]

$$\partial_\tau\chi_j = \boldsymbol{\nabla}_\xi\cdot[\mathsf{D}(\boldsymbol{\nabla}_\xi\chi_j + \boldsymbol{e}_j)], \quad \langle\boldsymbol{\nabla}_\xi\chi_j\rangle = 0, \qquad \mathsf{D} = \varepsilon\mathsf{I} + \mathsf{H}, \tag{A-39}$$

where $\langle\boldsymbol{\nabla}_\xi\chi_j\rangle = 0$ follows from the periodicity of χ_j. We stress that equation (A-37) involves the slow $(t, \boldsymbol{x})$ and fast variables $(\tau, \boldsymbol{\xi})$, while equation (A-39) involves only the fast variables. As the remainder of the analysis involves only the fast variables, for notational simplicity, we will drop the subscripts ξ shown in equation (A-39) and use ∂_t to denote ∂_τ.

We now recast the first formula in equation (A-39) in a more suggestive, divergence form. Define the operator $\mathbf{T} : \tilde{\mathcal{A}}_{\mathcal{T}} \to \mathcal{H}_{\mathcal{T}}$ by $(\mathbf{T}\boldsymbol{\psi})_j = \partial_t \psi_j$, $j = 1, \dots, d$. For $f \in \mathscr{F}$ we have [29, 74, 34]

$$\boldsymbol{\nabla}(-\Delta)^{-1}\partial_t f = (-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\nabla} f, \tag{A-40}$$

in a weak sense. This allows $\partial_t \chi_j$ in (A-39) to be written in divergence form [29, 74], $\partial_t \chi_j = (-\Delta)(-\Delta)^{-1}\partial_t \chi_j = -\boldsymbol{\nabla}\cdot[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]\boldsymbol{\nabla}\chi_j$. Define the vector-valued function $\boldsymbol{E}_j = \boldsymbol{\nabla}\chi_j + \boldsymbol{e}_j$ and the operator $\boldsymbol{\sigma} = \varepsilon \mathsf{I} + \mathbf{S}$, where $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$. With these definitions, the cell problem in (A-39) can be written via (A-36) as $\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{E}_j = 0$, $\langle \boldsymbol{E}_j \rangle = \boldsymbol{e}_j$, which is equivalent to

$$\boldsymbol{\nabla}\cdot\boldsymbol{J}_j = 0, \quad \boldsymbol{\nabla}\times\boldsymbol{E}_j = 0, \quad \boldsymbol{J}_j = \boldsymbol{\sigma}\boldsymbol{E}_j, \quad \langle \boldsymbol{E}_j \rangle = \boldsymbol{e}_j, \qquad \boldsymbol{\sigma} = \varepsilon \mathsf{I} + \mathbf{S}. \tag{A-41}$$

In the case of a time-independent fluid velocity field $\boldsymbol{u} = \boldsymbol{u}(\boldsymbol{x})$ we define $\mathbf{S} = \mathsf{H}$ and $\boldsymbol{\sigma} = \mathsf{D}$.

The formulas in (A-41) are analogous to the quasi-static limit of Maxwell's equations for a conductive medium [38, 68], where $\boldsymbol{E}_j$ and $\boldsymbol{J}_j$ play the role of the local electric field and current density, respectively, and $\boldsymbol{\sigma}$ plays the role of the local conductivity tensor of the medium. In the analytic continuation method for composites [38, 67, 10], the effective conductivity tensor $\boldsymbol{\sigma}^*$ is defined as

$$\langle \boldsymbol{J}_j \rangle = \boldsymbol{\sigma}^* \langle \boldsymbol{E}_j \rangle, \tag{A-42}$$

which relates the mean gradient field and flux. In the setting of a *time-independent* fluid velocity field, where $\mathbf{S} = \mathsf{H}$, the linear constitutive relation $\boldsymbol{J}_j = \boldsymbol{\sigma}\boldsymbol{E}_j$ in (A-41) relates the local gradient field and flux. In this case, due to the skew-symmetry of H, the local constitutive relationship is similar to that of a Hall medium [45, 29, 74, 68]. However, in the setting of a *time-dependent* fluid velocity field, where $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$, the constitutive relation $\boldsymbol{J}_j = \boldsymbol{\sigma}\boldsymbol{E}_j$ in (A-41) is a *non-local integro-differential equation.* The precise relationship between the bulk transport coefficients $\boldsymbol{\sigma}^*$ and D^* for the effective parameter problems of composite materials and advection-diffusion is addressed in Lemma 5 below.

We now derive functional formulas for the components S^*_{jk} and A^*_{jk}, $j, k = 1, \dots, d$, of the symmetric S^* and antisymmetric A^* parts of the effective diffusivity tensor D^* that are analogous to those shown in equation (15).

Writing the cell problem in (A-41) as $\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j = -\boldsymbol{\nabla}\cdot\mathsf{H}\boldsymbol{e}_j = -u_j$, and inserting this expression for u_j into the functional $\langle u_j\,\chi_k\rangle$ in (10) yields

$$\text{(A-43)}\qquad \begin{aligned}\langle u_j\,\chi_k\rangle &= -\langle[\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j]\,\chi_k\rangle\\ &= \langle\boldsymbol{\sigma}\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\nabla}\chi_k\rangle\\ &= \varepsilon\langle\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\nabla}\chi_k\rangle + \langle\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\nabla}\chi_k\rangle.\end{aligned}$$

Here, we have used the periodicity of χ_k and H in the second equality and the final equality follows from the property $\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j = \boldsymbol{\nabla}\chi_j$ and the symmetry of $\boldsymbol{\Gamma}$, together yielding $\langle\mathbf{S}\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\nabla}\chi_k\rangle = \langle\mathbf{S}\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_k\rangle = \langle\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j\cdot\boldsymbol{\nabla}\chi_k\rangle$. Equations (10), (11), and (A-43) imply that

$$\text{(A-44)}\qquad \mathsf{S}^*_{jk} = \varepsilon(\delta_{jk} + \langle\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times), \qquad \mathsf{A}^*_{jk} = \langle\mathbf{A}\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times, \qquad \mathbf{A} = \boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}.$$

We stress that $\boldsymbol{\Gamma}$ is a self-adjoint projection on $\mathcal{H}$, implying

$$\text{(A-45)}\qquad \langle\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times = \langle\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times = \langle\mathbf{S}\boldsymbol{\Gamma}\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times = \langle\mathbf{S}\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times.$$

Since $\boldsymbol{\nabla}\chi_k$ is real-valued we have $\langle\boldsymbol{\nabla}\chi_k,\boldsymbol{\nabla}\chi_j\rangle_\times = \langle\boldsymbol{\nabla}\chi_j,\boldsymbol{\nabla}\chi_k\rangle_\times$, implying that S^*, as defined by (A-44), is a symmetric matrix. By Young's inequality in (A-20), the operators ∂_t and $(-\Delta)^{-1}$ commute on $\tilde{\mathscr{A}}_{\mathcal{T}}\otimes\mathscr{H}_{\mathcal{V}}$ (see Theorem 2.27 in [34]). Therefore, we have $(-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\psi} = \mathbf{T}(-\boldsymbol{\Delta})^{-1}\boldsymbol{\psi}$, for $\boldsymbol{\psi}\in\mathcal{F}$ [34, 95]. This, the symmetry of $(-\boldsymbol{\Delta})^{-1}$ and the skew-symmetry of the operators $\mathbf{T}$ and H imply that the operator $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$ is skew-adjoint on $\mathcal{F}$. Since $\boldsymbol{\Gamma}$ is self-adjoint on $\mathcal{F}$, the operator $\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}$ is also skew-adjoint on $\mathcal{F}$. Just as in the discussion below equation (15), this implies that A^*, as defined by (A-44), is an antisymmetric matrix.

We stress that the operator $\boldsymbol{\Gamma}\mathsf{H} + (-\boldsymbol{\Delta})^{-1}\mathbf{T}$ is *unbounded* on the subspace of square-integrable gradients of spatio-temporal periodic functions in $\otimes_{j=1}^d L^2(\mathcal{T}\times\mathcal{V})$. This is contrary to the claim given in [56] while reviewing the formal results of [6], namely that the operator is compact on this function space. The unboundedness of the operator is due to the presence of the operator $\mathbf{T}$ which is unbounded on $\mathcal{H}_\mathcal{T}$. This can be understood by considering the orthonormal set of functions $\{\boldsymbol{\varphi}_n\}\subset\mathcal{H}_\mathcal{T}$ with components $(\boldsymbol{\varphi}_n)_j$, $j=1,\dots,d$, defined by $(\boldsymbol{\varphi}_n)_j(t) = \beta\sin((n+j)\pi t/T)$, $\beta = \sqrt{2/(Td)}$, satisfying $\langle\boldsymbol{\varphi}_n\cdot\boldsymbol{\varphi}_m\rangle_\mathcal{T} = \delta_{nm}$, which are analogous to the functions considered in (A-11). It follows from $(\partial_t\boldsymbol{\varphi}_n)_j(t) = [\beta(n+j)\pi/T]\cos((n+j)\pi t/T)$ and $\langle|\mathbf{T}\boldsymbol{\varphi}_n|^2\rangle_\mathcal{T} = \sum_j[(n+j)\pi/T]^2/d$ that the norm of $\mathbf{T}\boldsymbol{\varphi}_n$ grows arbitrarily large as $n \to \infty$. This establishes that $\mathbf{T}$ is unbounded on $\mathcal{H}_\mathcal{T}$. Consequently, in order to provide *rigorous* Stieltjes integral representations for D^* we must employ the spectral theory of *unbounded* self-adjoint operators.

Applying the integro-differential operator $\boldsymbol{\nabla}(-\Delta)^{-1}$ to the cell problem in equation (A-41), written via (A-36) as $\boldsymbol{\nabla\cdot\sigma\nabla}\chi_j = -\boldsymbol{\nabla}\cdot\mathsf{H}\boldsymbol{e}_j$, yields

$$\boldsymbol{\Gamma}(\varepsilon\mathsf{I}+\mathbf{S})\boldsymbol{\nabla}\chi_j = -\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j. \tag{A-46}$$

This and $\boldsymbol{\Gamma\nabla}\chi_j = \boldsymbol{\nabla}\chi_j$ provides the following resolvent formula for $\boldsymbol{\nabla}\chi_j$, which is analogous to equation (16),

$$\boldsymbol{\nabla}\chi_j = (\varepsilon\mathsf{I}+\mathbf{A})^{-1}\boldsymbol{g}_j, \qquad \boldsymbol{g}_j = -\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j. \tag{A-47}$$

Inserting the resolvent formula for $\boldsymbol{\nabla}\chi_j$ in equation (A-47) into (A-44) yields the following analogue of equation (17)

$$\begin{aligned} \mathsf{S}^*_{jk} &= \varepsilon\left(\delta_{jk} + \langle(\varepsilon\mathsf{I}+\mathbf{A})^{-1}\boldsymbol{g}_j, (\varepsilon\mathsf{I}+\mathbf{A})^{-1}\boldsymbol{g}_k\rangle_\times\right), \\ \mathsf{A}^*_{jk} &= \langle\mathbf{A}(\varepsilon\mathsf{I}+\mathbf{A})^{-1}\boldsymbol{g}_j, (\varepsilon\mathsf{I}+\mathbf{A})^{-1}\boldsymbol{g}_k\rangle_\times. \end{aligned} \tag{A-48}$$

The following corollary of Theorem 1 establishes the Stieltjes integral representations equation in (18) for the functional formulas of S^*_{jk} and A^*_{jk} in equation (A-48).

Corollary 4. *Assume $u_j \in \tilde{\mathscr{A}}_\mathcal{T}\otimes(\mathscr{H}^{0,2}_\mathcal{V}\cap L^\infty(\mathcal{V}))$ for all $j=1,\dots,d$. Then $\mathbf{A}=\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}$ is a maximal (skew-adjoint) normal operator on the function space $\mathcal{F}$, hence $\mathbf{M}=-\imath\mathbf{A}$ is self-adjoint on $\mathcal{F}$. Let $\mathbf{Q}(\lambda)$ be the resolution of the identity in one-to-one correspondence with $\mathbf{M}$. Define the complex valued function $\mu_{jk}(\lambda)=\langle\mathbf{Q}(\lambda)\boldsymbol{g}_j,\boldsymbol{g}_k\rangle_\times$, $j,k=1,\dots,d$, where $\boldsymbol{g}_j=-\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j$. Consider the positive measure μ_{kk} and the signed measures $Re\,\mu_{jk}$ and $Im\,\mu_{jk}$ associated with $\mu_{jk}(\lambda)$, introduced in equations (A-4) and (A-5). Then, for all $0<\varepsilon<\infty$, the functional formulas for S^*_{jk} and A^*_{jk} shown in (A-48) have the Radon–Stieltjes integral representations shown in equation (18).*

Proof of Corollary 4. We first establish that the operator $\mathbf{M}=-\imath\mathbf{A}$ with domain $\mathcal{F}$ is self-adjoint, where $\mathbf{A}=\boldsymbol{\Gamma}\mathbf{S}\boldsymbol{\Gamma}$ and $\mathbf{S}=(-\boldsymbol{\Delta})^{-1}\mathbf{T}+\mathsf{H}$. Let's first consider the operator $-\imath\boldsymbol{\Gamma}[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]\boldsymbol{\Gamma}$ with domain $\mathcal{F}$. Since $\boldsymbol{\Gamma}:\mathcal{H}_\mathcal{V}\to\mathcal{H}_\times$ is a projection, it acts as the identity on $\mathcal{H}_\times$. We can therefore focus on the operator $\imath[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]$. Since the function spaces $\mathscr{F}$ and $\mathcal{F}$ differ only in the characterization of the spatial variable, the one-to-one isometry $\mathscr{H}^{1,2}_\mathcal{V}\sim\mathcal{H}_\times$ established in Lemma 3 induces the one-to-one isometry $\mathscr{F}\sim\mathcal{F}$. Consequently, for all $\boldsymbol{\psi},\boldsymbol{\phi}\in\mathcal{F}$ there exist unique $f,h\in\mathscr{F}$ such that

$\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\boldsymbol{\phi} = \boldsymbol{\nabla} h$. Therefore, recalling that $\langle \boldsymbol{\psi}, \boldsymbol{\phi} \rangle_\times = \langle \boldsymbol{\psi} \boldsymbol{\cdot} \boldsymbol{\phi} \rangle$, by equation (A-40), we have

$$\text{(A-49)} \qquad \langle (-\boldsymbol{\Delta})^{-1} \mathbf{T} \boldsymbol{\psi}, \boldsymbol{\phi} \rangle_\times = \langle \boldsymbol{\nabla} (-\Delta)^{-1} \partial_t f \boldsymbol{\cdot} \boldsymbol{\nabla} h \rangle = \langle (-\Delta)^{-1} \partial_t f, h \rangle_{1,2}.$$

In the proof of Theorem 1 we established that the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F}$ is self-adjoint. This and equation (A-49) establishes that the operator $-\imath(-\boldsymbol{\Delta})^{-1}\mathbf{T}$ with domain $\mathcal{F}$ is self-adjoint.

Now focus on the operator $-\imath \boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma}$ with domain $\mathcal{H}$. Since $\boldsymbol{\Gamma}$ is a self-adjoint operator on $\mathcal{H}_\times$ and $-\imath \mathsf{H}$ is a Hermitian matrix, the operator $-\imath \boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma}$ is symmetric on $\mathcal{H}$. Recall that equation (A-35) provides the following representation of the fluid velocity field $\boldsymbol{u} = \boldsymbol{\nabla} \boldsymbol{\cdot} \mathsf{H}$. We now establish that $\boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma}$ is bounded on $\mathcal{H}$ when the components u_j, $j = 1, \dots, d$, of $\boldsymbol{u}$ satisfy $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V}))$. This, in turn, establishes that the operator $-\imath \boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma}$ with domain $\mathcal{H}$ is self-adjoint.

The one-to-one isometry $\mathscr{H}^{1,2}_{\mathcal{V}} \sim \mathcal{H}_\times$ established in Lemma 3 induces the one-to-one isometry $\mathscr{H} \sim \mathcal{H}$. Therefore, for every $\boldsymbol{\psi} \in \mathcal{H}$ there exists unique $f \in \mathscr{H}$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$. Consequently, since the operator $\boldsymbol{\Gamma} = -\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\boldsymbol{\cdot}$ acts as the identity on $\mathcal{H}_\times$ and $\boldsymbol{u} = \boldsymbol{\nabla} \boldsymbol{\cdot} \mathsf{H}$, equation (A-36) implies

$$\text{(A-50)} \quad \|\boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma} \boldsymbol{\psi}\|_\times = \|\boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\nabla} f\|_\times = \|\boldsymbol{\nabla}(-\Delta)^{-1}[\boldsymbol{u} \boldsymbol{\cdot} \boldsymbol{\nabla} f]\|_\times = \|(-\Delta)^{-1}[\boldsymbol{u} \boldsymbol{\cdot} \boldsymbol{\nabla} f]\|_{1,2}.$$

This, Lemma 2, and Lemma 3, in turn, show that $\boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma}$ is bounded on $\mathcal{H}$.

We now summarize our findings. We have established that the operator $-\imath \boldsymbol{\Gamma}[(-\boldsymbol{\Delta})^{-1}\mathbf{T}]\boldsymbol{\Gamma}$ with domain $\mathcal{F}$ is self-adjoint and the operator $-\imath \boldsymbol{\Gamma} \mathsf{H} \boldsymbol{\Gamma}$ with domain $\mathcal{H}$ is self-adjoint when the components u_j, $j = 1, \dots, d$, of $\boldsymbol{u}$ satisfy $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V}))$. Consequently, the sum of these two operators $\mathbf{M} = -\imath \mathbf{A}$, where $\mathbf{A} = \boldsymbol{\Gamma} \mathbf{S} \boldsymbol{\Gamma}$ and $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$, with domain $D(\mathbf{M}) = \mathcal{F} \cap \mathcal{H} = \mathcal{F}$ [97] is self-adjoint for $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}} \otimes (\mathscr{H}^{0,2}_{\mathcal{V}} \cap L^\infty(\mathcal{V}))$. Thus $\mathbf{A} = \imath \mathbf{M}$ is a maximal (skew-adjoint) normal operator on $\mathcal{F}$ [97].

In the proof of Theorem 1 we established that the functions $F(\lambda) = (\varepsilon + \imath\lambda)^{-1}$ and $G(\lambda) = \imath\lambda(\varepsilon + \imath\lambda)^{-1}$ involved in the functional formulas for S^*_{jk} and A^*_{jk} in (A-48) are bounded for all $0 < \varepsilon < \infty$ so that $\boldsymbol{\varphi} \in \mathscr{D}(F)$ and $\boldsymbol{\varphi} \in \mathscr{D}(G)$ for all $\boldsymbol{\varphi} \in D(\mathbf{M})$ when $0 < \varepsilon < \infty$. We now establish that $\boldsymbol{g}_j \in D(\mathbf{M})$. By equations (A-35) and (A-36), and the definition of $g_j = (-\Delta)^{-1} u_j$ in (16) we have

$$\text{(A-51)} \qquad \boldsymbol{g}_j = -\boldsymbol{\Gamma} \mathsf{H} \boldsymbol{e}_j = \boldsymbol{\nabla}(-\Delta)^{-1} u_j = \boldsymbol{\nabla} g_j.$$

In the proof of Theorem 1 we established that $g_j \in \mathscr{F}$. This, equation (A-51) and the correspondence $\mathscr{F} \sim \mathcal{F}$ via Lemma 3, imply that $\boldsymbol{g}_j \in \mathcal{F}$. Since $\mathcal{F} \subseteq D(\mathbf{M})$ (by construction), the conditions of the spectral theorem are satisfied. Just as in the remainder of the proof of Theorem 1, this establishes the integral representations for S^*_{jk} and A^*_{jk} shown in (18). From equation (A-7), the mass μ^0_{jk} of the measure μ_{jk} is given by

$$\mu^0_{jk} = \langle \boldsymbol{g}_j, \boldsymbol{g}_k \rangle_\times = \langle \boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j, \boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_k \rangle_\times = \langle \mathsf{H}^T\boldsymbol{\Gamma}\mathsf{H}\boldsymbol{e}_j, \boldsymbol{e}_k \rangle_\times . \tag{A-52}$$

Moreover, $|\mu^0_{jk}| \leq \|\mathsf{H}\|^2_\times < \infty$ for all $j, k = 1, \ldots, d$, where $\|\cdot\|_\times$ denotes the operator norm on $\mathcal{H}$. This completes the proof of Corollary 4. □

We conclude this section with the following lemma, which provides a precise relationship between the effective parameter $\boldsymbol{\sigma}^*$ defined in equation (A-42) and the effective parameter D^* defined in (10). This generalizes an analogous result given in [29].

Lemma 5. *Let the components D^*_{jk} and σ^*_{jk}, $j, k = 1, \ldots, d$, of the effective tensors D^* and $\boldsymbol{\sigma}^*$ be defined as in equations (4)–(10) and (A-37)–(A-42), respectively. Then these effective tensors are related by*

$$\boldsymbol{\sigma}^* = [\mathsf{D}^*]^T + \langle \mathsf{H} \rangle . \tag{A-53}$$

Proof of Lemma 5. Below equation (A-31) we discussed the the orthogonal decomposition $\mathcal{H}_\mathcal{V} = \mathcal{H}_\times \oplus \mathcal{H}_\bullet \oplus \mathcal{H}_0$. Temporally denote $\mathcal{F} = \tilde{\mathcal{A}}_\mathcal{T} \otimes \mathcal{H}_\times$ by $\mathcal{F}_\times$ and define the function space $\mathcal{F}_\bullet = \tilde{\mathcal{A}}_\mathcal{T} \otimes \mathcal{H}_\bullet$. From equation (A-41), the vector-valued functions $\boldsymbol{J}_j = \boldsymbol{\sigma}\boldsymbol{E}_j$ and $\boldsymbol{E}_j = \boldsymbol{\nabla}\chi_j + \boldsymbol{e}_j$ satisfy $\boldsymbol{J}_j \in \mathcal{F}_\bullet$ and $\boldsymbol{E}_j \in \mathcal{F}_\times \oplus \mathcal{H}_0$ while $\boldsymbol{\nabla}\chi_j \in \mathcal{F}_\times$, where $\boldsymbol{\sigma} = \varepsilon\mathsf{I} + \mathbf{S}$ and $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$. By the mutual orthogonality of the Hilbert spaces $\mathcal{H}_\times$ and $\mathcal{H}_\bullet$ and the Fubini-Tonelli theorem [34] we have $\langle \boldsymbol{J}_j \cdot \boldsymbol{\nabla}\chi_k \rangle = 0$ for all $j, k = 1, \ldots, d$ (which is equivalent to equation (A-43)). Consequently, from equations (A-41) and (A-42) we have $\langle \boldsymbol{J}_j \cdot \boldsymbol{E}_k \rangle = \langle \boldsymbol{J}_j \cdot \boldsymbol{e}_k \rangle = \sigma^*_{jk}$.

The definition $\boldsymbol{u} = \boldsymbol{\nabla} \cdot \mathsf{H}$ in (A-35), periodicity, and integration by parts yields $\langle \boldsymbol{\nabla}\chi_j \cdot \mathsf{H}\boldsymbol{e}_k \rangle = -\langle \chi_j \, u_k \rangle$. From $\mathbf{S} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \mathsf{H}$ we have $\mathbf{S}\boldsymbol{e}_j = \mathsf{H}\boldsymbol{e}_j$. Therefore, the skew-symmetry of $\mathbf{S}$, the mean-zero property $\langle \boldsymbol{\nabla}\chi_j \rangle = 0$, and the formula $\mathsf{D}^*_{jk} = \varepsilon\delta_{jk} + \langle u_j \chi_k \rangle$ in (10) yield

$$\begin{aligned} \sigma^*_{jk} &= \langle (\varepsilon\mathsf{I} + \mathbf{S})(\boldsymbol{\nabla}\chi_j + \boldsymbol{e}_j) \cdot \boldsymbol{e}_k \rangle \\ &= \langle (\varepsilon\mathsf{I} + \mathbf{S})\boldsymbol{\nabla}\chi_j \cdot \boldsymbol{e}_k \rangle + \langle (\varepsilon\mathsf{I} + \mathbf{S})\boldsymbol{e}_j \cdot \boldsymbol{e}_k \rangle \\ &= -\langle \boldsymbol{\nabla}\chi_j \cdot \mathsf{H}\boldsymbol{e}_k \rangle + \langle (\varepsilon\mathsf{I} + \mathsf{H})\boldsymbol{e}_j \cdot \boldsymbol{e}_k \rangle \end{aligned} \tag{A-54}$$

$$= \langle \chi_j \, u_k \rangle + \varepsilon \delta_{jk} + \langle \mathsf{H}_{jk} \rangle$$
$$= \mathsf{D}^*_{kj} + \langle \mathsf{H}_{jk} \rangle,$$

which is equivalent to (A-53). This concludes our proof of Lemma 5. □

Appendix D. An isometric correspondence

In this section we show that the effective parameter problem described in Theorem 1 is equivalent to the effective parameter problem described in Corollary 4. The correspondence between the two formulations is one of isometry, and is summarized by the following theorem.

Theorem 6. *The function spaces* $\mathscr{F}$ *and* $\mathcal{F}$ *defined in equations* (A-16) *and* (A-33) *are in one-to-one isometric correspondence. This induces a one-to-one isometric correspondence between the domains* $D(A)$ *and* $D(\mathbf{A})$ *of the operators* A *and* $\mathbf{A}$ *defined in equations* (15) *and* (A-44)*, respectively. Specifically,* $\mathscr{F} \subseteq D(A)$ *and* $\mathcal{F} \subseteq D(\mathbf{A})$*. Moreover, for every* $f \in \mathscr{F}$ *we have* $\boldsymbol{\nabla} f \in \mathcal{F}$ *and* $\|Af\|_{1,2} = \|\mathbf{A}\boldsymbol{\nabla} f\|_\times$*. Conversely, for each* $\boldsymbol{\psi} \in \mathcal{F}$ *there exists unique* $f \in \mathscr{F}$ *such that* $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ *and* $\|\mathbf{A}\boldsymbol{\psi}\|_\times = \|Af\|_{1,2}$*. The Radon–Stieltjes measures underlying the integral representations of Theorem 1 and Corollary 4 are equal,* $\mathrm{d}\langle Q(\lambda) g_j, g_k \rangle_{1,2} = \mathrm{d}\langle \mathbf{Q}(\lambda) \boldsymbol{g}_j, \boldsymbol{g}_k \rangle_\times$*,* $j, k = 1, \ldots, d$*, up to null sets of measure zero, where* $\boldsymbol{g}_j = \boldsymbol{\nabla} g_j$*. Moreover, the operators* $\mathbf{A}$ *and* A *are related by* $\mathbf{A}\boldsymbol{\nabla} = \boldsymbol{\nabla} A$*, which implies and is implied by the weak equality* $\mathbf{Q}(\lambda)\boldsymbol{\nabla} = \boldsymbol{\nabla} Q(\lambda)$*.*

Proof of Theorem 6. Recall, we have $\boldsymbol{\nabla} g_j = \boldsymbol{g}_j$ from equation (A-51). We use the formula $\boldsymbol{u} = \boldsymbol{\nabla}\cdot\mathsf{H}$ in equation (A-35) and the weak identity in (A-36) to write the operator $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$ defined in (15) as $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{\nabla}\cdot\mathsf{H}\boldsymbol{\nabla})$. Using the definition $\boldsymbol{\Gamma} = -\boldsymbol{\nabla}(-\Delta)^{-1}\boldsymbol{\nabla}\cdot$ in (A-32), the formula $\boldsymbol{\nabla}(-\Delta)^{-1}\partial_t = (-\boldsymbol{\Delta})^{-1}\mathbf{T}\boldsymbol{\nabla}$ in (A-40), and the representation $\mathbf{A} = (-\boldsymbol{\Delta})^{-1}\mathbf{T} + \boldsymbol{\Gamma}\mathsf{H}$, which holds in the weak sense shown in (A-45), the operators A and $\mathbf{A}$ are related by

$$\boldsymbol{\nabla} A = [(-\boldsymbol{\Delta})^{-1}\mathbf{T} + \boldsymbol{\Gamma}\mathsf{H}]\boldsymbol{\nabla} = \mathbf{A}\boldsymbol{\nabla}, \qquad \boldsymbol{\nabla} g_j = \boldsymbol{g}_j. \tag{A-55}$$

Consequently, by applying the differential operator $\boldsymbol{\nabla}$ to both sides of the formula $(\varepsilon + A)\chi_j = g_j$ of (16), we obtain the formula $(\varepsilon\mathsf{I} + \mathbf{A})\boldsymbol{\nabla}\chi_j = \boldsymbol{g}_j$ of equation (A-47).

As discussed in the proof of Corollary 4, the one-to-one isometry $\mathscr{H}^{1,2}_{\mathcal{V}} \sim \mathcal{H}_\times$ established in Lemma 3 induces the one-to-one isometry $\mathscr{F} \sim \mathcal{F}$. By construction, in Appendices C.1 and C.2 we established that $\mathscr{F} \subseteq D(A)$ and $\mathcal{F} \subseteq D(\mathbf{A})$, respectively. Therefore, the one-to-one isometry between these subsets $\mathscr{F}$ and $\mathcal{F}$ of $D(A)$ and $D(\mathbf{A})$, respectively, induces a one-to-one isometric correspondence between these subsets of the domains $D(A)$ and $D(\mathbf{A})$ of the operators A and $\mathbf{A}$, respectively. We will make the relationship between the domains $D(A)$ and $D(\mathbf{A})$ more precise below. We now show that this isometric correspondence and the one-to-one correspondence between a self-adjoint operator and its resolution of the identity, discussed in the paragraph containing (A-10), establishes that the mathematical frameworks given in Appendices C.1 and C.2 produce equivalent Stieltjes integral representations for the effective diffusivity tensor D^*.

In Appendix A we discussed that the domain $D(M)$ of the self-adjoint operator M comprises those and only those elements f of $\mathscr{H}$ such that the Stieltjes integral $\int \lambda^2\, \mathrm{d}\|Q(\lambda)f\|_{1,2}^2$ is convergent, and when $f \in D(M)$ the element Mf is determined by the relations in equation (A-10), with suitable notational changes. Since $A = \imath M$ it is clear that $D(A) = D(M)$. Analogous statements hold for the self-adjoint operator $\mathbf{M}$.

Let $f \in \mathscr{F}$. The relation $\mathscr{F} \sim \mathcal{F}$ implies $\boldsymbol{\nabla} f \in \mathcal{F}$, so from (A-55)

$$\|Af\|_{1,2}^2 = \langle Af, Af\rangle_{1,2} = \langle \boldsymbol{\nabla} Af \cdot \boldsymbol{\nabla} Af\rangle = \langle \mathbf{A}\boldsymbol{\nabla} f \cdot \mathbf{A}\boldsymbol{\nabla} f\rangle = \|\mathbf{A}\boldsymbol{\nabla} f\|_{\times}^2. \tag{A-56}$$

Consequently, from equation (A-10) we have

$$\int_{-\infty}^{\infty} \lambda^2\, \mathrm{d}\|Q(\lambda)f\|_{1,2}^2 = \int_{-\infty}^{\infty} \lambda^2\, \mathrm{d}\|\mathbf{Q}(\lambda)\boldsymbol{\nabla} f\|_{\times}^2, \tag{A-57}$$

and the convergence of the integral on the left side of (A-57) implies the convergence of the integral on the right side of (A-57). This, in turn, implies $\boldsymbol{\nabla} f \in D(\mathbf{A})$.

Conversely, let $\boldsymbol{\psi} \in \mathcal{F}$. From the relation $\mathscr{F} \sim \mathcal{F}$, there exists unique $f \in \mathscr{F}$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$. Equation (A-55) then implies

$$\|\mathbf{A}\boldsymbol{\psi}\|_{\times}^2 = \langle \mathbf{A}\boldsymbol{\nabla} f, \mathbf{A}\boldsymbol{\nabla} f\rangle_{\times} = \langle \boldsymbol{\nabla} Af, \boldsymbol{\nabla} Af\rangle_{\times} = \langle Af, Af\rangle_{1,2} = \|Af\|_{1,2}^2. \tag{A-58}$$

Again, equation (A-10) implies that (A-57) holds, and the convergence of the integral on the right side of (A-57) implies the convergence of the integral on the left side of (A-57) which, in turn, implies that $f \in D(A)$.

In summary, for every $f \in \mathscr{F}$ we have $\boldsymbol{\nabla} f \in D(\mathbf{A})$ and $\|Af\|_{1,2}^2 = \|\mathbf{A}\boldsymbol{\nabla} f\|_{\times}^2$. Conversely, for every $\boldsymbol{\psi} \in \mathcal{F}$, there exists unique $f \in D(A)$ such that $\boldsymbol{\psi} = \boldsymbol{\nabla} f$ and $\|\mathbf{A}\boldsymbol{\psi}\|_{\times}^2 = \|Af\|_{1,2}^2$. This generates a one-to-one isometric correspondence between the domains $D(\mathbf{A})$ and $D(A)$.

We now show that this result implies, and is implied by the weak equality $\boldsymbol{\nabla} Q(\lambda) = \mathbf{Q}(\lambda)\boldsymbol{\nabla}$, where $Q(\lambda)$ and $\mathbf{Q}(\lambda)$ are the *self-adjoint projection operators* in one-to-one correspondence with the operators A and $\mathbf{A}$, respectively. From equation (A-57) and the linearity properties of Radon–Stieltjes integrals [97], we have that

$$0 = \int_{-\infty}^{\infty} \lambda^2 \,\mathrm{d}(\|Q(\lambda)f\|_{1,2}^2 - \|\mathbf{Q}(\lambda)\boldsymbol{\nabla} f\|_{\times}^2) = \int_{-\infty}^{\infty} \lambda^2 \,\mathrm{d}(\langle[\boldsymbol{\nabla} Q(\lambda) - \mathbf{Q}(\lambda)\boldsymbol{\nabla}]f \boldsymbol{\cdot} \boldsymbol{\nabla} f\rangle). \tag{A-59}$$

Equation (A-59) implies that $\mathrm{d}\|Q(\lambda)f\|_{1,2}^2 = \mathrm{d}\|\mathbf{Q}(\lambda)\boldsymbol{\nabla} f\|_{\times}^2$, up to sets of measure zero, for all $f \in \mathscr{F} \iff \boldsymbol{\nabla} f \in \mathcal{F}$, and $\boldsymbol{\nabla} Q(\lambda) = \mathbf{Q}(\lambda)\boldsymbol{\nabla}$ in this weak sense. Conversely, assume that $Q(\lambda)$ and $\mathbf{Q}(\lambda)$ are the resolutions of the identity in one-to-one correspondence with the operators A and $\mathbf{A}$ and that $\boldsymbol{\nabla} Q(\lambda)f = \mathbf{Q}(\lambda)\boldsymbol{\nabla} f$ for every $f \in \mathscr{F} \iff \boldsymbol{\nabla} f \in \mathcal{F}$. Then equation (A-59) holds and implies equation (A-57). Equation (A-10) then implies that $\|\mathbf{A}\boldsymbol{\nabla} f\|_{\times}^2 = \|Af\|_{1,2}^2 = \|\boldsymbol{\nabla} Af\|_{\times}^2$, which implies that $\mathbf{A}\boldsymbol{\nabla} = \boldsymbol{\nabla} A$ in this weak sense. Since $g_k \in D(A)$ and $\boldsymbol{g}_k \in D(\mathbf{A})$ with $\boldsymbol{g}_k = \boldsymbol{\nabla} g_k$, this result implies that the Radon–Stieltjes measures underlying the integral representations of Theorem 1 are equal to the measures of Corollary 4, $\mathrm{d}\langle Q(\lambda)g_j, g_k\rangle_{1,2} = \mathrm{d}\langle \mathbf{Q}(\lambda)\boldsymbol{g}_j, \boldsymbol{g}_k\rangle_{\times}$, up to null sets of measure zero, for all $j, k = 1, \ldots, d$. This concludes our proof of Theorem 6. □

Appendix E. Discrete integral representations by eigenfunction expansion

The integral representations of Theorem 1 and Corollary 4 shown in equation (18), involve a Stieltjes measure μ_{jk}, $j, k = 1, \ldots, d$, that has discrete and continuous components [88, 97]. In this section, we review these properties of μ_{jk} and provide an explicit formula for its discrete component. Towards this goal, in Appendix E.1 we summarize some general spectral properties of the self-adjoint operators $M = -\imath A$ and $\mathbf{M} = -\imath\mathbf{A}$ on the function spaces $\mathscr{F}$ and $\mathcal{F}$, which are dense subsets of the associated Hilbert spaces $\mathscr{H}$ and $\mathcal{H}$, given in equations (A-16) and (A-33), respectively. We will focus on the operator M and the Hilbert space $\mathscr{H}$, as the discussion regarding $\mathbf{M}$ and $\mathcal{H}$ is analogous. In Appendix E.2 we refine the result in Appendix E.1, applying it to the space of fluid velocity fields that have finite (trigonometric) Fourier series.

E.1. General methods

Recall from equation (A-10) that the domain $D(M)$ of the self-adjoint operator M comprises those and only those elements $f \in \mathscr{H}$ such that $\|Mf\|_{1,2}^2 = \int_{-\infty}^{\infty} \lambda^2 \, \mathrm{d}\|Q(\lambda)f\|_{1,2}^2 < \infty$, where $Q(\lambda)$ is the resolution of the identity in one-to-one correspondence with M [97]. The integration is over the spectrum Σ of M, which has continuous Σ_{cont} and discrete (pure-point) Σ_{pp} components, $\Sigma = \Sigma_{\mathrm{cont}} \cup \Sigma_{\mathrm{pp}}$ [88, 97]. We first focus on the discrete spectrum Σ_{pp}.

The $f \in \mathscr{H}$, $f \neq 0$, satisfying $Mf = \lambda f$ with $\lambda \in \Sigma_{\mathrm{pp}}$ are called eigenfunctions and λ is the corresponding eigenvalue. Since M is self-adjoint, λ is real-valued [97]. The span of all eigenfunctions is a *countable* subspace of $\mathscr{H}$ [97]. Accordingly, we will denote the eigenfunctions by φ_l, $l = 1, 2, 3, \ldots$, with corresponding eigenvalues λ_l. Eigenfunctions corresponding to distinct eigenvalues are orthogonal and can be normalized to be orthonormal [97], i.e. if $M\varphi_l = \lambda_l \varphi_l$ and $M\varphi_m = \lambda_m \varphi_m$ for $\lambda_l \neq \lambda_m$, then $\langle \varphi_m, \varphi_n \rangle_{1,2} = \delta_{mn}$. There can be more than one eigenfunction associated with a particular eigenvalue. However, they are linearly independent and, without loss of generality, can be taken to be orthonormal [97]. Consequently, associated with each eigenfunction φ_l is a closed linear manifold, which we denote by $\mathcal{M}(\varphi_l)$. When $l \neq m$, $\mathcal{M}(\varphi_l)$ and $\mathcal{M}(\varphi_m)$ are mutually orthogonal.

Set $\mathcal{E} = \oplus_{l=1}^{\infty} \mathcal{M}(\varphi_l)$, $\mathcal{M} = \mathcal{E} \oplus \{0\}$, and let $\mathcal{N} = \mathcal{M}^{\perp}$ be the orthogonal complement of $\mathcal{M}$ in $\mathscr{H}$. There exists a countable orthonormal basis $\{\psi_m\}$ for $\mathcal{N}$ [97]. Denote by $\mathcal{N}(\psi_m)$ the closed linear manifolds associated with each basis element ψ_m of $\mathcal{N}$.

All the properties of $\mathcal{M}$ and $\mathcal{N}$ that are relevant here have been collected in the following theorem [97], which provides a natural decomposition of the Hilbert space $\mathscr{H}$ in terms of the mutually orthogonal, closed linear manifolds $\mathcal{M}$ and $\mathcal{N}$, and leads to a decomposition of the measure μ_{kk} into its discrete and continuous components.

Theorem 7 ([97] pages 189 and 247)**.** *One of the three cases must occur:*

1. *$\mathcal{E} = \emptyset$ and $\mathcal{M} = \{0\}$ has dimension zero; $\mathcal{N} = \mathscr{H}$ has countably infinite dimension. There exists an orthonormal set $\{\psi_m\}$, $m = 1, 2, 3, \ldots$, and mutually orthogonal, closed linear manifolds $\mathcal{N}(\psi_m)$ which determine $\mathcal{N}$ according to $\mathcal{N} = \oplus_{m=1}^{\infty} \mathcal{N}(\psi_m)$.*
2. *$\mathcal{E}$ contains an incomplete orthonormal set $\{\varphi_l\}$ so that both $\mathcal{M}$ and $\mathcal{N}$ are proper subsets of $\mathscr{H}$, $\mathcal{N}$ having countably infinite dimension and $\mathcal{M}$ having finite or countably infinite dimension. There exists an orthonormal set $\{\psi_m\}$ in $\mathcal{N}$. The closed linear manifolds $\mathcal{M}(\varphi_l)$ and*

$\mathcal{N}(\psi_m)$ are mutually orthogonal and together determine $\mathscr{H}$ according to

$$\mathcal{M} = \oplus_{l=1}^{\infty}\mathcal{M}(\varphi_l), \qquad \mathcal{N} = \oplus_{m=1}^{\infty}\mathcal{N}(\psi_m), \qquad \mathscr{H} = \mathcal{M} \oplus \mathcal{N}.$$

3. *$\mathcal{E}$ contains a complete orthonormal set $\{\varphi_l\}$; $\mathcal{M} = \mathscr{H}$ has countably infinite dimension; $\mathcal{N} = \{0\}$ has zero dimension. In this case, the closed linear manifolds $\mathcal{M}(\varphi_l)$ are mutually orthogonal and together determine $\mathcal{M}$ according to $\mathcal{M} = \oplus_{l=1}^{\infty}\mathcal{M}(\varphi_l)$.*

In each of these three cases, the closed linear manifolds $\mathcal{M}$ and $\mathcal{N}$ reduce M, i.e., M leaves both $\mathcal{M}$ and $\mathcal{N}$ invariant in the sense that if $f \in D(M)$ and $f \in \mathcal{N}$ then $Mf \in \mathcal{N}$, and similarly for $\mathcal{M}$. In cases (2) and (3), a necessary and sufficient condition that an element $\varphi_l \in \mathscr{H}$ is an eigenfunction with eigenvalue λ_l, is that the function $\|Q(\lambda)\varphi_l\|_{1,2}^2$ is constant on each of the intervals $-\infty < \lambda < \lambda_l$ and $\lambda_l < \lambda < \infty$ [97]. Moreover, a necessary and sufficient condition that $f \in \mathcal{M}$, $f \neq 0$, is

$$f = \sum_{l=1}^{\infty}\langle f, \varphi_l\rangle_{1,2}\,\varphi_l, \qquad \|f\|_{1,2}^2 = \sum_{l=1}^{\infty}|\langle f, \varphi_l\rangle_{1,2}|^2 \neq 0, \tag{A-60}$$

and similarly for $f \in \mathcal{N}$ with orthonormal set $\{\psi_m\}$. In cases (1) and (2), a necessary and sufficient condition that $\psi \neq 0$ be an element of $\mathcal{N}$ is that $\|Q(\lambda)\psi\|_{1,2}^2$ be a continuous function of λ not identically zero [97].

Let f be an arbitrary element of $\mathscr{H}$, and g and h be its (unique [34]) projections on $\mathcal{M}$ and $\mathcal{N}$, respectively, then the equation

$$\begin{aligned} \|Q(\lambda)f\|_{1,2}^2 &= \|Q(\lambda)g\|_{1,2}^2 + \|Q(\lambda)h\|_{1,2}^2, \\ \mathrm{d}\|Q(\lambda)f\|_{1,2}^2 &= \mathrm{d}\|Q(\lambda)g\|_{1,2}^2 + \mathrm{d}\|Q(\lambda)h\|_{1,2}^2, \end{aligned} \tag{A-61}$$

is valid and provides the standard decomposition of the monotone function $\|Q(\lambda)f\|_{1,2}^2$ into its discontinuous and continuous monotone components, as well as the decomposition of the measure $\mathrm{d}\|Q(\lambda)f\|_{1,2}^2$ into its discrete and continuous components.

We now use the mathematical framework summarized in Theorem 7 to provide explicit formulas for the discrete parts of the integral representations for S_{jk}^* and A_{jk}^*, shown in equation (18). Recall the cell problem in equation (9) written as in (A-17), $(\varepsilon + A)\chi_j = g_j$. Here, $A = \imath M$ is defined in (15), $g_j = (-\Delta)^{-1}u_j$, and u_j is the j^{th} component of the velocity field $\boldsymbol{u}$, $j = 1, \ldots, d$. Moreover, by Theorem 1 we have $\chi_j, g_j \in \mathscr{F} \subset \mathscr{H}$ and

$\mathscr{F} \subseteq D(A)$. We emphasize that the arguments presented here are more subtle than those typically used for *bounded* operators in Hilbert space. The reason is a bounded linear operator typically commutes with all the infinite sums encountered here, by the dominated convergence theorem [34]. However, for the operator A, we must instead rely on general principles of unbounded linear operators in Hilbert space.

Let $\tilde{\chi}_j$ and $\chi_j^{\perp}$ be the (unique) projections of χ_j on $\mathcal{M}$ and $\mathcal{N}$, respectively, with $\chi_j = \tilde{\chi}_j + \chi_j^{\perp}$ and similarly for g_j. Since $A = \imath M$ is a linear operator, we have $A\chi_j = A\tilde{\chi}_j + A\chi_j^{\perp}$. From Theorem 7, the linear manifolds $\mathcal{M}$ and $\mathcal{N}$ both reduce A, which implies $A\tilde{\chi}_j \in \mathcal{M}$ and $A\chi_j^{\perp} \in \mathcal{N}$. From equation (A-60) we then have $A\tilde{\chi}_j = \sum_l \langle A\tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l$ and

$$\chi_j = \sum_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l + \chi_j^{\perp}, \quad A\chi_j = \sum_l \imath\lambda_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l + A\chi_j^{\perp}, \tag{A-62}$$

where $\langle A\tilde{\chi}_j, \varphi_l\rangle_{1,2} = -\langle \tilde{\chi}_j, A\varphi_l\rangle_{1,2} = -\langle \tilde{\chi}_j, \imath\lambda_l\varphi_l\rangle_{1,2} = \imath\lambda_l\langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}$ was used. From the cell problem $(\varepsilon + A)\chi_j = g_j$ we therefore have

$$\varepsilon \sum_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l + \sum_l \imath\lambda_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \varphi_l + (\varepsilon + A)\chi_j^{\perp} = \tilde{g}_j + g_j^{\perp}, \tag{A-63}$$

where $(\varepsilon + A)\chi_j^{\perp}, g_j^{\perp} \in \mathcal{N}$. Of course, each $f \in \mathcal{N}$ can be represented [97] as $f = \sum_m \langle f, \psi_m\rangle_{1,2}\, \psi_m$, where $\{\psi_m\}$ is the orthonormal set defined in Theorem 7, though we have suppressed this notation in the above equations for simplicity. By the mutual orthogonality of the linear manifolds $\mathcal{M}$ and $\mathcal{N}$, the completeness of the set $\{\varphi_l\} \cup \{\psi_m\}$, and Parseval's identity [90], taking the inner-product of both sides of equation (A-63) with φ_n yields

$$\langle \tilde{\chi}_j, \varphi_n\rangle_{1,2} = \frac{\langle \tilde{g}_j, \varphi_n\rangle_{1,2}}{\varepsilon + \imath\lambda_n}\,, \qquad 0 < \varepsilon < \infty. \tag{A-64}$$

Recall the functional representations $\mathsf{S}^*_{jk} = \varepsilon(\delta_{jk} + \langle \chi_j, \chi_k\rangle_{1,2})$ and $\mathsf{A}^*_{jk} = \langle A\chi_j, \chi_k\rangle_{1,2}$, $j, k = 1, \ldots, d$, in equation (15). Writing $\chi_j = \tilde{\chi}_j + \chi_j^{\perp}$ and $A\chi_j = A\tilde{\chi}_j + A\chi_j^{\perp}$, the mutual orthogonality of the linear manifolds $\mathcal{M}$ and $\mathcal{N}$, which both reduce A, implies $\langle \chi_j, \chi_k\rangle_{1,2} = \langle \tilde{\chi}_j, \tilde{\chi}_k\rangle_{1,2} + \langle \chi_j^{\perp}, \chi_k^{\perp}\rangle_{1,2}$ and $\langle A\chi_j, \chi_k\rangle_{1,2} = \langle A\tilde{\chi}_j, \tilde{\chi}_k\rangle_{1,2} + \langle A\chi_j^{\perp}, \chi_k^{\perp}\rangle_{1,2}$. Consequently, from equations (A-62) and (A-64), the completeness of the set $\{\varphi_l\} \cup \{\psi_m\}$, and Parseval's identity [90], we have

$$\langle \chi_j, \chi_k\rangle_{1,2} - \langle \chi_j^{\perp}, \chi_k^{\perp}\rangle_{1,2} = \sum_l \langle \tilde{\chi}_j, \varphi_l\rangle_{1,2}\, \overline{\langle \tilde{\chi}_k, \varphi_l\rangle}_{1,2} \tag{A-65}$$

$$
\begin{aligned}
&= \sum_l \frac{\langle \tilde{g}_j, \varphi_l \rangle_{1,2} \, \overline{\langle \tilde{g}_k, \varphi_l \rangle}_{1,2}}{\varepsilon^2 + \lambda_l^2} \\
\langle A\chi_j, \chi_k \rangle_{1,2} - \langle A\chi_j^\perp, \chi_k^\perp \rangle_{1,2} &= \sum_l \imath\lambda_l \, \langle \tilde{\chi}_j, \varphi_l \rangle_{1,2} \, \overline{\langle \tilde{\chi}_k, \varphi_l \rangle}_{1,2} \\
&= \sum_l \frac{\imath\lambda_l \, \langle \tilde{g}_j, \varphi_l \rangle_{1,2} \, \overline{\langle \tilde{g}_k, \varphi_l \rangle}_{1,2}}{\varepsilon^2 + \lambda_l^2} \, .
\end{aligned}
$$

Since χ_j and $A\chi_j$ are real-valued, just as in equation (A-9), we have

$$
\begin{aligned}
\langle \chi_j, \chi_k \rangle_{1,2} - \langle \chi_j^\perp, \chi_k^\perp \rangle_{1,2} &= \sum_l \frac{\mathrm{Re}\,[\langle \tilde{g}_j, \varphi_l \rangle_{1,2} \, \overline{\langle \tilde{g}_k, \varphi_l \rangle}_{1,2}]}{\varepsilon^2 + \lambda_l^2} \\
\langle A\chi_j, \chi_k \rangle_{1,2} - \langle A\chi_j^\perp, \chi_k^\perp \rangle_{1,2} &= -\sum_l \frac{\lambda_l \, \mathrm{Im}\,[\langle \tilde{g}_j, \varphi_l \rangle_{1,2} \, \overline{\langle \tilde{g}_k, \varphi_l \rangle}_{1,2}]}{\varepsilon^2 + \lambda_l^2} \, .
\end{aligned} \tag{A-66}
$$

The right sides of the formulas in equation (A-66) are Radon–Stieltjes integrals associated with a *discrete* measure. Equation (A-66) establishes the series representations shown in equation (19).

The terms $\langle \chi_j^\perp, \chi_k^\perp \rangle_{1,2}$ and $\langle A\chi_j^\perp, \chi_k^\perp \rangle_{1,2}$ also have Radon–Stieltjes integral representations involving the *continuous* measure $\mathrm{d}\langle Q(\lambda) g_j^\perp, g_k^\perp \rangle_{1,2}$ via equation (18). We note that from the decomposition $g_j = \tilde{g}_j + g_j^\perp$, we have $\langle \tilde{g}_j, \varphi_l \rangle_{1,2} = \langle g_j, \varphi_l \rangle_{1,2}$. A useful property of the inner-product $\langle g_j, \varphi_l \rangle_{1,2}$ and the form of $g_j = (-\Delta)^{-1} u_j$ is (see equation (A-21))

$$
\langle g_j, \varphi_l \rangle_{1,2} = \langle u_j, \varphi_l \rangle, \tag{A-67}
$$

where $\langle \cdot, \cdot \rangle$ is the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$–inner-product defined below equation (A-14). This property is used in Appendix E.2 to calculate S_{jk}^* and A_{jk}^* for a large class of fluid velocity fields.

E.2. Trigonometric Fourier methods

In this section we refine the results shown in equations (A-66) and (A-67), applying them to the class of fluid velocity fields $\boldsymbol{u}$ that have components u_j, $j = 1, \ldots, d$, which are representable by *finite* trigonometric Fourier series. For the sake of notational simplicity and correspondence with Sections 3 and 4 we set $\mathcal{T} \times \mathcal{V} = [0, 2\pi]^{d+1}$. Let $\boldsymbol{u} \in \mathcal{U}_N$, where $\mathcal{U}_N = \otimes_{j=1}^d \mathcal{U}_N^j$ and

$$
\mathcal{U}_N^j = \left\{ f_j \;\middle|\; f_j = \sum_{(\ell, \boldsymbol{k}) \in \mathbb{Z}_N^{d+1}} b_{\ell, \boldsymbol{k}}^j \, \phi_{\ell, \boldsymbol{k}} \right\}, \quad b_{\ell, \boldsymbol{k}}^j = \big\langle f_j, \phi_{\ell, \boldsymbol{k}} \big\rangle. \tag{A-68}
$$

Here, $\mathbb{Z}_N^n = \{\boldsymbol{q} \in \mathbb{Z}^n \mid -N \le q_i \le N,\ i = 1, \ldots, n,\ N \in \mathbb{N}\}$, $\langle \cdot, \cdot \rangle$ denotes the *sesquilinear* $\mathscr{H}_{\mathcal{TV}}$–inner-product defined below equation (A-14) and $\phi_{\ell,\boldsymbol{k}}(t, \boldsymbol{x}) = \exp[\imath(\ell t + \boldsymbol{k}\cdot\boldsymbol{x})]$. In the discussion in Appendix C.1.2 we established that $\langle u_j \rangle_{\mathcal{V}} = 0$, which implies $\langle u_j \rangle = 0$, but it is not necessary that $\langle u_j \rangle_{\mathcal{T}} = 0$. Therefore, the sum $u_j = \sum_{\ell,\boldsymbol{k}} b^j_{\ell,\boldsymbol{k}} \phi_{\ell,\boldsymbol{k}}$ runs over the index set $I_N = \{(\ell, \boldsymbol{k}) \in \mathbb{Z}_N^{d+1} \mid \boldsymbol{k} \neq 0\}$ when $\langle u_j \rangle_{\mathcal{T}} \neq 0$ and $I_N = \{(\ell, \boldsymbol{k}) \in \mathbb{Z}_N^{d+1} \mid \ell \neq 0,\ \boldsymbol{k} \neq 0\}$ when $\langle u_j \rangle_{\mathcal{T}} = 0$ as well. Our class $\mathcal{U}_N$ of fluid velocity fields is broad, as $\{\phi_{\ell,\boldsymbol{k}} \mid (\ell, \boldsymbol{k}) \in \mathbb{Z}^{d+1}\}$ is a *complete* orthonormal basis for $\mathscr{H}_{\mathcal{TV}}$ [34], implying the set $\cup_{N<\infty} \mathcal{U}_N$ is dense in $\mathscr{H}_{\mathcal{TV}}$ [90].

Consider the eigenvalue problem $A\varphi_l = \imath\lambda_l\varphi_l$, $\lambda_l \in \mathbb{R}$, $l \in \mathbb{N}$, involving the integro-differential operator $A = (-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})$ defined in (15)

$$(-\Delta)^{-1}(\partial_t - \boldsymbol{u}\cdot\boldsymbol{\nabla})\varphi_l = \imath\lambda_l\varphi_l. \tag{A-69}$$

From $\varphi_l \in \mathscr{F}$, $\mathscr{F} = \tilde{\mathscr{A}}_{\mathcal{T}} \otimes \mathscr{H}^{1,2}_{\mathcal{V}}$, equation (A-22), and the definition of $\tilde{\mathscr{A}}_{\mathcal{T}}$ in (A-13), we have $(-\Delta)^{-1}\partial_t\varphi_l \in \mathscr{H}_{\mathcal{T}} \otimes \mathscr{H}^{1,2}_{\mathcal{V}}$. By Lemma 2 and $\varphi_l \in \mathscr{F}$ we have $(-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]\varphi_l \in \mathscr{F}$. Consequently, both of these functions are members of the Hilbert space $\mathscr{H}_{\mathcal{TV}}$. Since $\{\phi_{\ell,\boldsymbol{k}} \mid (\ell, \boldsymbol{k}) \in \mathbb{Z}^{d+1}\}$ is a complete orthonormal basis for $\mathscr{H}_{\mathcal{TV}}$ [34], we have for $(\ell, \boldsymbol{k}) \in \mathbb{Z}^{d+1}$

$$\begin{gathered}\varphi_l = \sum_{\ell,\boldsymbol{k}} \langle \varphi_l, \phi_{\ell,\boldsymbol{k}} \rangle\, \phi_{\ell,\boldsymbol{k}}\,, \quad (-\Delta)^{-1}\partial_t\varphi_l = \sum_{\ell,\boldsymbol{k}} \left\langle (-\Delta)^{-1}\partial_t\varphi_l, \phi_{\ell,\boldsymbol{k}} \right\rangle \phi_{\ell,\boldsymbol{k}}\,, \\ (-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]\varphi_l = \sum_{\ell,\boldsymbol{k}} \left\langle (-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]\varphi_l, \phi_{\ell,\boldsymbol{k}} \right\rangle \phi_{\ell,\boldsymbol{k}}\,. \end{gathered} \tag{A-70}$$

We now identify the index set for the series in equation (A-70). In the discussion in Appendices C.1.1 and C.1.2 we established the domain of the operator $(-\Delta)^{-1}$ is contained in the space $\mathscr{H}_{\mathcal{V}} \backslash \mathbb{C}^d$ of $\mathcal{V}$–periodic functions that are also spatially mean-zero. Moreover, when f is in this domain then $(-\Delta)^{-1}f$ is also spatially mean-zero, $\langle (-\Delta)^{-1}f \rangle_{\mathcal{V}} = 0$. Since $\varphi_l \in \mathscr{F}$ it follows that $\langle \varphi_l \rangle_{\mathcal{V}} = 0$, thus $\langle (-\Delta)^{-1}\partial_t\varphi_l \rangle_{\mathcal{V}} = \partial_t\langle (-\Delta)^{-1}\varphi_l \rangle_{\mathcal{V}} = 0$ (see Theorem 2.27 in [34]). Since, $\boldsymbol{\nabla}\cdot\boldsymbol{u} = 0$ (weakly), we have $\boldsymbol{u}\cdot\boldsymbol{\nabla}\varphi_l = \boldsymbol{\nabla}\cdot(\boldsymbol{u}\,\varphi_l)$. Therefore, by the divergence theorem [63] and spatial periodicity, the function $\boldsymbol{u}\cdot\boldsymbol{\nabla}\varphi_l$ is spatially mean-zero, thus $\langle (-\Delta)^{-1}[\boldsymbol{u}\cdot\boldsymbol{\nabla}]\varphi_l \rangle_{\mathcal{V}} = 0$. Consequently, the series in equation (A-70) run over the index set $I = \{(\ell, \boldsymbol{k}) \in \mathbb{Z}^{d+1} \mid \boldsymbol{k} \neq 0\}$.

Since the orthonormal basis $\{\phi_{\ell,\boldsymbol{k}} \mid (\ell, \boldsymbol{k}) \in \mathbb{Z}^{d+1}\}$ is complete in $\mathscr{H}_{\mathcal{TV}}$, the Fourier series representation of $A\varphi_l \in \mathscr{H}_{\mathcal{TV}}$ converges in norm topology no matter how the series is ordered [34]. Moreover, by the completeness of the basis $\sum_{\ell,\boldsymbol{k}} c_{\ell,\boldsymbol{k}}\,\phi_{\ell,\boldsymbol{k}} = 0$ only if $c_{\ell,\boldsymbol{k}} = 0$ for all $(\ell,\,\boldsymbol{k}) \in I$ [34]. Consequently, plugging the formulas in (A-70) into equation (A-69) yields

$$\text{(A-71)}\quad \left\langle (-\Delta)^{-1}\partial_t\varphi_l, \phi_{\ell,\boldsymbol{k}}\right\rangle - \left\langle (-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]\varphi_l, \phi_{\ell,\boldsymbol{k}}\right\rangle - \imath\lambda_l\,\langle\varphi_l, \phi_{\ell,\boldsymbol{k}}\rangle = 0.$$

It is clear that $\partial_t\phi_{\ell,\boldsymbol{k}} = \imath\ell\,\phi_{\ell,\boldsymbol{k}}$ and $\boldsymbol{\nabla}\phi_{\ell,\boldsymbol{k}} = \imath\boldsymbol{k}\,\phi_{\ell,\boldsymbol{k}}$. Since, for all $t\in\mathcal{T}$, $\phi_{\ell,\boldsymbol{k}}(t,\cdot)\in C^\infty(\mathcal{V})$ and $-\Delta\phi_{\ell,\boldsymbol{k}} = |\boldsymbol{k}|^2\phi_{\ell,\boldsymbol{k}}$, applying $(-\Delta)^{-1}$ to both sides of this formula yields $(-\Delta)^{-1}\phi_{\ell,\boldsymbol{k}} = |\boldsymbol{k}|^{-2}\phi_{\ell,\boldsymbol{k}}$ (see Theorem 1 in Section 4.2 of [63]). In the proof of Theorem 1 in Appendix C.1.3 we established that the operator $-\imath(-\Delta)^{-1}\partial_t$ with domain $\mathscr{F}$ is self-adjoint and the operator $-\imath(-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ with domain $\mathscr{H}$ is self-adjoint when $u_j \in \tilde{\mathscr{A}}_{\mathcal{T}}\otimes(\mathscr{H}^{0,2}_{\mathcal{V}}\cap L^\infty(\mathcal{V}))$ for all $j = 1,\dots,d$, which is clearly satisfied for $\boldsymbol{u}\in\mathcal{U}^N$. Consequently, since the operators $(-\Delta)^{-1}$ and ∂_t are symmetric and skew-adjoint in the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$–inner-product, respectively, we have

$$\text{(A-72)}\qquad \left\langle (-\Delta)^{-1}\partial_t\varphi_l, \phi_{\ell,\boldsymbol{k}}\right\rangle = \left\langle \varphi_l, -\imath\ell|\boldsymbol{k}|^{-2}\phi_{\ell,\boldsymbol{k}}\right\rangle, = \imath\ell|\boldsymbol{k}|^{-2}\left\langle \varphi_l, \phi_{\ell,\boldsymbol{k}}\right\rangle.$$

Moreover, the calculation in (A-29) shows the operator $[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]$ is skew-adjoint in the $\mathscr{H}_{\mathcal{T}\mathcal{V}}$–inner-product. Therefore, denoting $u_j = \sum_{\ell',\boldsymbol{k}'} b^j_{\ell',\boldsymbol{k}'}\phi_{\ell',\boldsymbol{k}'}$ and $\boldsymbol{b}_{\ell',\boldsymbol{k}'} = \big(b^1_{\ell',\boldsymbol{k}'},\dots,b^d_{\ell',\boldsymbol{k}'}\big)$, we have

$$\text{(A-73)}\qquad \begin{aligned}\left\langle (-\Delta)^{-1}[\boldsymbol{u}\boldsymbol{\cdot}\boldsymbol{\nabla}]\varphi_l, \phi_{\ell,\boldsymbol{k}}\right\rangle &= \left\langle \varphi_l, -[\boldsymbol{u}\boldsymbol{\cdot}\,\imath\boldsymbol{k}]|\boldsymbol{k}|^{-2}\phi_{\ell,\boldsymbol{k}}\right\rangle\\ &= \sum_{\ell',\boldsymbol{k}'} \imath[\overline{\boldsymbol{b}_{\ell',\boldsymbol{k}'}}\boldsymbol{\cdot}\boldsymbol{k}]|\boldsymbol{k}|^{-2}\left\langle \varphi_l, \phi_{\ell+\ell',\boldsymbol{k}+\boldsymbol{k}'}\right\rangle,\end{aligned}$$

where $\phi_{\ell,\boldsymbol{k}}\,\phi_{\ell',\boldsymbol{k}'} = \phi_{\ell+\ell',\boldsymbol{k}+\boldsymbol{k}'}$, $(\ell',\boldsymbol{k}')\in I_N$, and $(\ell,\boldsymbol{k})\in I$. Inserting equations (A-72) and (A-73) into equation (A-71), removing the common factor of $\imath$, and denoting $a^l_{\ell,\boldsymbol{k}} = \langle\varphi_l, \phi_{\ell,\boldsymbol{k}}\rangle$ yields the following Fourier representation of the eigenvalue problem $A\varphi_l = \imath\lambda_l\varphi_l$

$$\text{(A-74)}\qquad |\boldsymbol{k}|^{-2}\Big(\ell\,a^l_{\ell,\boldsymbol{k}} - \sum_{\ell',\boldsymbol{k}'}[\overline{\boldsymbol{b}_{\ell',\boldsymbol{k}'}}\boldsymbol{\cdot}\boldsymbol{k}]\,a^l_{\ell+\ell',\boldsymbol{k}+\boldsymbol{k}'}\Big) = \lambda_l\,a^l_{\ell,\boldsymbol{k}}.$$

Equation (A-74) is an infinite system of algebraic equations that determines the eigenvalues λ_l and Fourier coefficients $a^l_{\ell,\boldsymbol{k}}$ of the eigenfunctions φ_l of the self-adjoint operator $M = -\imath A$.

The Fourier representation of the spectral weights $\langle g_j,\varphi_l\rangle_{1,2}\,\overline{\langle g_k,\varphi_l\rangle}_{1,2}$ in (A-66) are determined as follows. Since $\{\phi_{\ell,\boldsymbol{k}} \mid (\ell,\boldsymbol{k})\in\mathbb{Z}^{d+1}\}$ is a *complete* *orthonormal* basis for $\mathscr{H}_{\mathcal{TV}}$, equations (A-67), (A-68), (A-70), and Parseval's identity [90] imply

$$\text{(A-75)} \qquad \langle g_j, \varphi_l \rangle_{1,2} = \langle u_j, \varphi_l \rangle = \sum_{\ell', \boldsymbol{k}'} b^j_{\ell', \boldsymbol{k}'} \, \overline{a^l_{\ell', \boldsymbol{k}'}} \,, \qquad (\ell', \boldsymbol{k}') \in I_N \,.$$

Parseval's identity [90] also implies the Fourier representation of the orthogonality relation $\langle \boldsymbol{\nabla}\varphi_l \cdot \boldsymbol{\nabla}\varphi_m \rangle = \delta_{lm}$ is

$$\text{(A-76)} \qquad \delta_{lm} = \langle \boldsymbol{\nabla}\varphi_l \cdot \boldsymbol{\nabla}\varphi_m \rangle = \sum_{\ell, \boldsymbol{k}} |\boldsymbol{k}|^2 \, a^l_{\ell, \boldsymbol{k}} \, \overline{a^m_{\ell, \boldsymbol{k}}} \,, \qquad (\ell, \boldsymbol{k}) \in I.$$

Truncating the index set for $(\ell, \boldsymbol{k})$ in equation (A-74) defines an eigenvalue problem $\mathsf{C}^{-1}\mathsf{B}\boldsymbol{a}_l = \lambda_l \boldsymbol{a}_l$, involving a diagonal matrix C with values $|\boldsymbol{k}|^2$ along its diagonal and a matrix B that is Hermitian, as the fluid velocity field $\boldsymbol{u}$ is real-valued which implies the terms $\boldsymbol{b}_{\ell', \boldsymbol{k}'}$ in its Fourier series come in complex conjugate pairs. This can be written as the generalized eigenvalue problem $\mathsf{B}\boldsymbol{a}_l = \lambda_l \mathsf{C}\boldsymbol{a}_l$. However, in general, $|\boldsymbol{k}|^2 \, a^l_{\ell, \boldsymbol{k}}$ does not have a finite limit as $|\boldsymbol{k}| \to \infty$. This generalized eigenvalue problem can be rewritten as the standard eigenvalue problem $[\mathsf{C}^{-1/2}\mathsf{B}\mathsf{C}^{-1/2}][\mathsf{C}^{1/2}\boldsymbol{a}_l] = \lambda_l[\mathsf{C}^{1/2}\boldsymbol{a}_l]$, which is defined even for the infinite system via equation (A-76). This standard eigenvalue problem is used in Sections 3 and 4 to compute the discrete part of the spectral measure and integral representation of the effective diffusivity for the velocity field in (1).

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