The O–H bond strength of 83 kcal mol^{-1} in $HMnO_4^-$ is a critical value because it is well established that rates of radical reactions are directly related to bond strengths. For main-group radicals such as $(CH_3)CO\cdot$ ($^tBuO\cdot$), the Polanyi equation (*23*) states that the activation barrier for an H· transfer reaction, or log *k*, is proportional to the driving force, $\Delta H°$. The $\Delta H°$ for a H· transfer reaction $A{-}H + B \rightleftarrows A + H{-}B$ is simply the difference between the A–H and B–H bond strengths. An ·OH abstracts a benzylic hydrogen from toluene at an almost diffusion-limited rate because H· transfer is highly exothermic (31 kcal mol^{-1}), based on the bond dissociation energies $D(PhCH_2{-}H) = 88.5 \pm 1.5$ kcal mol^{-1} and $D(HO{-}H) = 119.3$ kcal mol^{-1} (*24*). Reactions of toluene with $^tBuO\cdot$ and especially $^tBuOO\cdot$ are slower because the reactions are less favorable, in the last case, downhill by only 1 kcal mol^{-1} [$D(^tBuO{-}H) = 104.8$ kcal mol^{-1}, $D(^tBuOO{-}H) = 89.4$ kcal mol^{-1} (*24*)].

A plot of the Polanyi relation, log *k* versus $\Delta H°$ for H· abstraction from toluene, is shown in Fig. 2. The rate of H· abstraction by MnO_4^- correlates with the rates of reaction of main-group radicals. In other words, MnO_4^- abstracts H· from toluene just as an oxygen radical would if it had a bond strength of 83 kcal mol^{-1}. This is true even though MnO_4^- is not a radical. Similar correlations have been found for oxidations of ethylbenzene by RO· and MnO_4^- (*6*) and for oxidations of toluene, cyclohexane, and isobutane by RO· and CrO_2Cl_2 [on the basis of an estimated CrO–H bond strength (*18*, *19*)]. The rate of toluene oxidation by MnO_4^- in water does not fit this relation because the mechanism is not H· abstraction; rates of H· transfer are essentially independent of solvent (*25*).

The correlation of rate with driving force provides insight into how a metal-containing active site can remove a hydrogen atom from an organic substrate. This correlation should apply not only to reagents such as permanganate but also to active sites in metalloenzymes and on metal oxide surfaces. The key feature of such an active site is not its radical character but its affinity for H·. Understanding these reactions should be based primarily on the thermochemistry of the H· abstraction step.

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Global Distribution of Persistent Organochlorine Compounds

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The global distribution of 22 potentially harmful organochlorine compounds was investigated in more than 200 tree bark samples from 90 sites worldwide. High concentrations of organochlorines were found not only in some developing countries but also in industrialized countries, which continue to be highly contaminated even though the use of many of these compounds is restricted. The distribution of relatively volatile organochlorine compounds (such as hexachlorobenzene) is dependent on latitude and demonstrates the global distillation effect, whereas less volatile organochlorine compounds (such as endosulfan) are not as effectively distilled and tend to remain in the region of use.

Researchers have long speculated that some organic pollutants move through the atmosphere from relatively warm source regions and condense at colder, higher latitudes onto vegetation, soil, and bodies of water. This process, known as the global distillation effect, could be the cause of the high concentrations of some pollutants found in Earth's arctic regions (*1–5*). Global distillation is driven by the change of a pollutant's subcooled liquid vapor pressure with temperature, its environmental persistence, and its tendency to associate with lipids (*1*, *6*). This effect appears to be most pronounced for organochlorine compounds of intermediate volatility, many of which are used as insecticides and fungicides. Some of these compounds are still in use throughout the world (for example, endosulfan), whereas others have been banned in almost all developed countries (for example, DDT). Understanding the environmental fate of these compounds is particularly important because some of them are carcinogens and some may be estrogen mimics (*7*).

These ubiquitous organochlorine pollutants have subcooled liquid vapor pressures of 0.1 to 0.001 Pa at 25°C (*8*). Although these vapor pressures are sufficient for all of

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these compounds to be transported globally to some degree, the most volatile compounds would be more readily distilled. Thus the more volatile organochlorine compounds, such as hexachlorobenzene (HCB) and the hexachlorocyclohexanes (HCHs), would be expected to occur in relatively higher concentrations at higher latitudes. Less volatile compounds such as endosulfan and DDT would not be as effectively transported to high latitudes, and their environmental concentrations would tend to reflect their local usage.

Because of the difficulty of collecting samples from all over the world, researchers have primarily attempted to study the global distillation effect on regional scales (*2*, *3*) and by the use of global models (*6*). Calamari *et al.* were among the first to study this effect on a global scale; they analyzed leaf, lichen, and moss samples from 26 sites worldwide (*4*). The distributions of HCB and lower chlorinated polychlorinated biphenyls have been observed to correlate with latitude or ambient temperature in several studies (*1*, *2*, *4*), whereas the distribution of other volatile organochlorine compounds, such as the HCHs, did not (*1–4*). Chernyak *et al.* recently reported that α-HCH concentrations in seawater increased with latitude on a transect from the Sea of Japan to the Bering Sea, but a similar trend was not observed for γ-HCH (*3*).

Unfortunately, any attempt to investigate global distillation of organochlorine compounds is complicated by a general lack of data about their sources. Although attempts have been made to compile local usage data for some organochlorine compounds (*9*), only rough estimates for a limited number of countries are available (*10*). Our goal was to determine the global distribution of organochlorine compounds with the use of samples collected from many sites worldwide. By creating a large data set, we hoped to determine the status of organochlorine contamination in industrialized countries as well as in developing countries. We also hoped to identify which organochlorine compounds show strong evidence of global distillation and which compounds tend to remain near their region of use. We measured 22 organochlorine compounds, including active ingredients, metabolites,

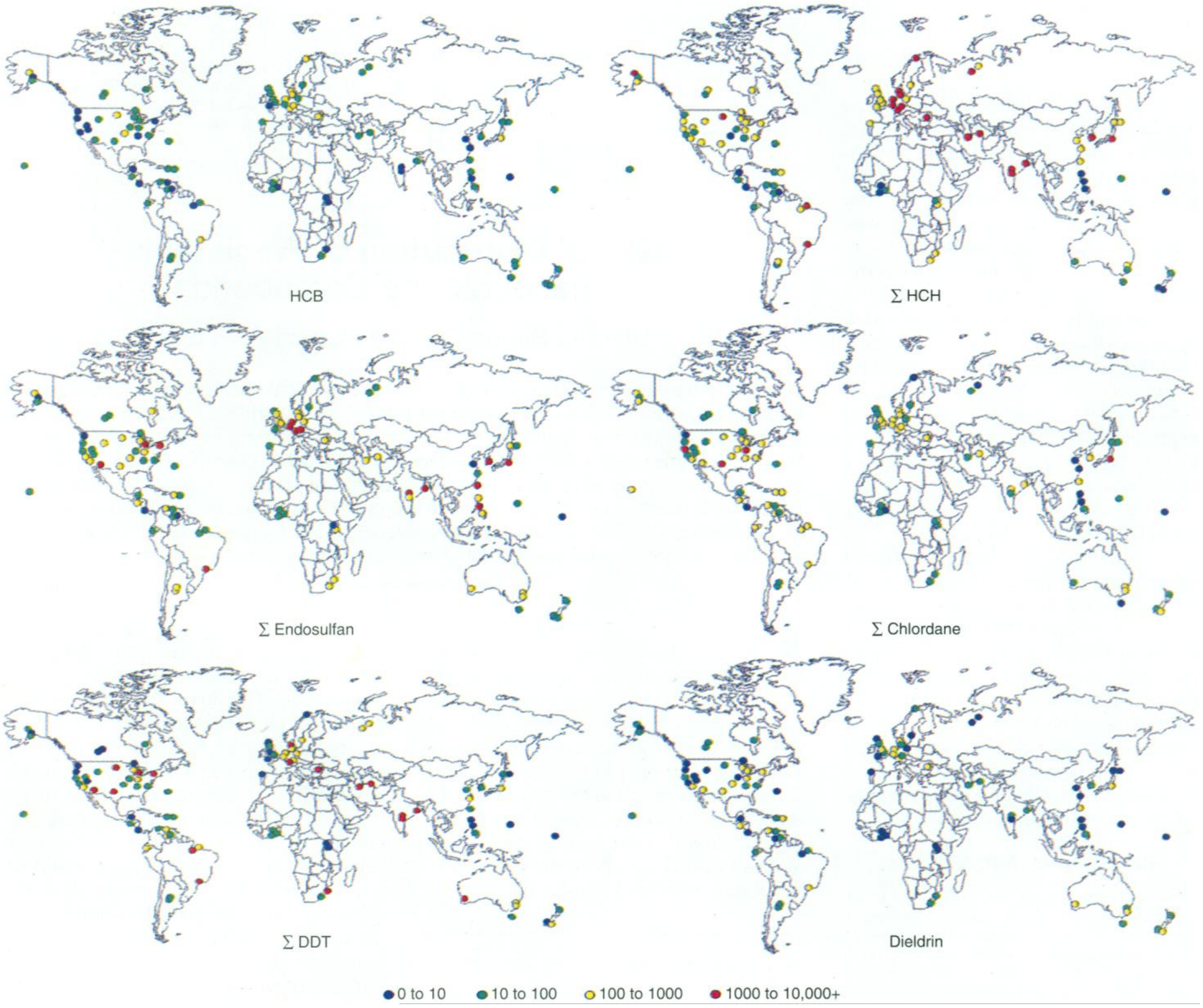

Fig. 1. Concentrations of organochlorine compounds (in nanograms of organochlorine per gram of lipid) measured at 90 sites in 209 tree bark samples (Σ endosulfan, sum of endosulfan I, endosulfan II, and endosulfan sulfate; Σ DDT, sum of *p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT; Σ HCH, sum of α-, β-, γ-, and δ-HCH; and Σ chlordane, sum of oxychlordane, heptachlor epoxide, *trans*-chlordane, *cis*-chlordane, and *trans*-nonachlor). At most sites, two or more samples were analyzed; in these cases, the concentrations were averaged. Tabular data are available from the authors.

and degradation products. The compounds included four HCHs, HCB, pentachloroanisole (PCA), dieldrin, three endosulfans, six chlordanes, two endrins, aldrin, and *p,p'*-DDT (and its degradation products *p,p'*-DDE and *p,p'*-DDD).

More than 200 tree bark samples were collected from 90 sites worldwide between 1992 and 1995 (Fig. 1) (*11*). Bark is an excellent scavenger of lipophilic organic pollutants from the atmosphere, even at low atmospheric concentrations (*12*), as a result of its surface area and associated lipids (*13*, *14*). Tree bark is present throughout the world, and on average it contains more lipids per unit area than other forms of vegetation (*13*, *14*). In addition, bark remains on most trees for several years, thus integrating lipophilic organic pollutants over an extended time. Because bark was collected from many different species of trees worldwide, we normalized the measured organochlorine concentrations to the bark's lipid concentration (in nanograms of organochlorine per gram of lipid). Because these compounds are sorbed to the bark surface from the atmosphere and do not undergo translocation, this normalization tends to account for variability attributable to differences in tree species (*13*, *14*).

Samples were fortified with isotopically labeled internal standards and extracted with solvent. The extracts were purified on silica gel columns (*15*) and analyzed by electron-capture gas chromatographic mass spectrometry on a Hewlett-Packard 5989A instrument (*16*). Moisture and lipid content were determined as in (*14*). Moisture content ranged from 2 to 70% by weight and lipids ranged from 0.2 to 3% by weight. The relative standard deviation of the measurements from nearby sites ranged from 4 to 80% of the measured average organochlorine concentration, depending on the compound and proximity to nearby sources.

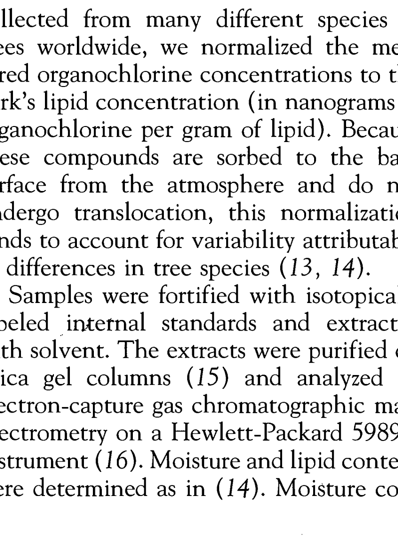

Fig. 2. Logarithms of concentrations (in nanograms of organochlorine per gram of lipid) of HCB (**A**) and endosulfan II (**B**) in tree bark are plotted against the absolute value of the latitude where the tree bark sample was taken. The regression lines are $a_0 = 0.927$, $a_1 = 0.0138$, $r = 0.482$, $n = 90$ (in A) and $a_0 = 1.18$, $a_1 = 0.0017$, $r = 0.033$, $n = 90$ (in B).

As shown in Fig. 1, almost all of the organochlorines measured in this study are ubiquitous on a global scale except for heptachlor, aldrin, and the two endrins, all of which degrade to other compounds (*17*). Low but measurable organochlorine concentrations were found even in tree bark samples from remote regions of the world, including the Orinoco rain forest in Venezuela, Guanacaste National Park in Costa Rica, and the rain forests of Ecuador and Belize. Tree bark samples from remote islands, including the Marshall Islands, Guam, Bermuda, Tasmania, and the South Island of New Zealand, also had low but measurable concentrations of organochlorine compounds. Samples from Uganda, Togo, and Ghana contained low concentrations of contaminants.

High concentrations of organochlorine compounds were measured in tree bark from the United States, Europe, India, the Middle East, Japan, Brazil, Australia, Taiwan, South Korea, and Russia. Samples from agricultural areas of the midwestern and eastern United States showed high concentrations of contaminants, as did samples from agricultural areas in California and the southwestern United States. In Europe, contamination was consistently high in parts of Germany, Romania, Belgium, the Netherlands, and Denmark. In Japan, contamination was consistently high in and near Tokyo and lower in Hokkaido.

The organochlorine compounds found in highest concentrations were the HCHs, the endosulfans, and *p,p'*-DDE (a degradation product of *p,p'*-DDT). The HCHs were primarily found as the α- and γ-isomers and are used as insecticides either as the pure γ-isomer (commonly known as lindane) or as a technical mixture of different isomers. Although the technical mixture is restricted in most developed countries, lindane is not (*10*). Ratios of α-HCH to γ-HCH concentration in bark were greater than 2 in parts of the United States, China, India, and Japan, which indicates past or present use of the technical HCH mixture. High concentrations of HCHs were measured in samples from Norway, Alaska, Canada, Sweden, Scotland, Russia, and Hokkaido, Japan. All these sites showed low concentrations for most other organochlorine compounds. HCH concentrations were relatively low in the Philippines, Venezuela, Ecuador, Central America, Ghana, Togo, the Marshall Islands, and Guam. A similar pattern was observed for HCB, although the global concentrations were much lower (Fig. 1).

Endosulfan is one of the last hexachlorocyclopentadiene insecticides remaining in widespread use (*18*), and it was found in high concentrations throughout the world (Fig. 1). Endosulfan is used on most crops, but the high concentrations of endosulfan measured in India and the Pacific Rim may result from its intensive use in rice cultivation (*18*). It is clear from Fig. 1 that industrialized countries are making full use of this readily available insecticide.

The insecticide *p,p'*-DDT was found in high concentrations relative to its degradation product, *p,p'*-DDE, in samples from India, Iran, Romania, Russia, and the west coast of Australia. The median ratio of *p,p'*-DDT to *p,p'*-DDE concentration in bark was greater than 2 in these countries, which indicates possible recent use of this insecticide. Even though DDT has been banned in the United States since 1973, its degradation products are very persistent in the environment; high concentrations of *p,p'*-DDE were still found in parts of the Midwest and Southwest.

In our analysis of global distillation, we used the absolute value of the latitude of the sampling sites as the independent variable. The organochlorine concentrations measured at each of 90 sites were found to have a log-normal distribution; thus, the concentrations were all logarithmically transformed and used as the dependent variable. We used a simple regression model,

$$\log\,[\mathrm{oc}] = a_0 + a_1\,|\mathrm{latitude}| \qquad (1)$$

where [oc] is the organochlorine concentration in the bark (in nanograms per gram of lipid) and |latitude| is the absolute value of the latitude of the sampling sites (in degrees). This model gave a significant relation ($P < 0.0001$) for α- and γ-HCH, HCB, and (to a lesser extent) PCA, the four most volatile compounds measured in this study (*19*). The slopes (a_1) were all positive, indicating that as latitude increases, the concentration in the bark increases. The concentration of HCB, a relatively volatile compound, is low in tree bark from equatorial regions compared with that in boreal and arctic regions (Fig. 2A). Less volatile compounds, such as endosulfan II (Fig. 2B), showed no significant correlation with latitude, which indicates that these compounds are not as effectively distilled and tend to remain near the original region of use. The distribution of these less volatile compounds is more likely related to the socioeconomic conditions of a given country (*20*, *21*) or to the types of crops grown there (*21*).

We conclude that relatively volatile organochlorine compounds (such as HCB and the HCHs) are readily distilled to colder, higher latitudes but that less volatile compounds (such as endosulfan and DDT) are not. Several factors appear to control the global distribution of these less volatile compounds: (i) chemical persistence in industrialized countries (such as the United States) where large amounts of these compounds were used in the past, (ii) present use in developing countries (such as India) where there is a market for inexpensive organochlorine compounds, and (iii) lack of use in countries (such as Ghana) that cannot afford to import and manufacture these compounds.

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11. Remote or rural sites were generally chosen for sampling, and permission was obtained from land owners, park rangers, or other officials. Samples were collected as described in (*14*). A 5 cm by 20 cm area of outer bark was chiseled off the tree, wrapped in aluminum foil, and stored in a plastic bag. Two to three sites were usually sampled at the same time, and three to six samples were collected from different species of trees at each site. A permit for importing tree bark samples into the United States was obtained from the U.S. Department of Agriculture, Animal and Plant Health Inspections Services. Samples were shipped by various worldwide carriers and were stored at –18°C until extraction.
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15. Unground tree bark (wet weight, 30 to 50 g) was spiked with an appropriate amount of six isotopically labeled internal standards (d_6-γ-HCH, $^{37}Cl_6$-heptachlor epoxide, d_4-endosulfan I, $^{37}Cl_6$-*trans*-nonachlor, d_8-*p,p′*-DDE, and d_8-*p,p′*-DDT) and Soxhlet-extracted in 50% hexane in acetone for 24 hours. This procedure extracted the organochlorine compounds from the bark. All extracts were solvent-exchanged to hexane and back-extracted 2 to 10 times with high-performance liquid chromatography–grade water until the water fraction was not turbid. This procedure removed polar interferents and improved the subsequent silica gel separation. Extracts were then reduced to 1 ml and purified with a silica gel column (length 25 cm, deactivated with 1% water by weight). The compounds were eluted from the column with hexane (50 ml), 50% hexane in CH_2Cl_2 (75 ml), and CH_2Cl_2 (75 ml). HCB eluted in the hexane fraction, endosulfan II and endosulfan cyclic sulfate eluted in the CH_2Cl_2 fraction, and most of the other compounds eluted in the 50% hexane in CH_2Cl_2 fraction. All fractions were combined for analysis. Recoveries of known amounts of organochlorine compounds were 76 to 84% for the entire procedure. Reagent blanks were done with every other set of extractions, and the blank experiments were free of contamination.
16. The gas chromatograph (GC) was equipped with a 30 m by 250 μm inside diameter (film thickness 0.25 μm) DB-5 fused silica capillary column (J&W Scientific, Folsom, CA). The ion source was maintained at a temperature of 125°C and a CH_4 pressure of 0.43 torr (direct source reading). After concentration to 200 μl under a gentle stream of N_2, 1 μl of the sample was injected splitless for 0.9 min. The GC temperature program conditions were 40°C held for 1 min, ramped at 30°C/min to 130°C, then at 3°C/min to 241°C, and finally at 30°C/min to 285°C and held for 10 min. The GC was set for a constant head pressure of 15 psi. The combination of these GC conditions and the selected ion monitoring program allowed us to analyze all 22 compounds in one injection. The detection limit ranged from 2 to 500 pg, depending on the electron-capture cross section of the analyte.
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22. We thank more than 70 scientists, friends, and colleagues who helped in the collection of samples.

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Magnetic Resonance Elastography by Direct Visualization of Propagating Acoustic Strain Waves

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A nuclear magnetic resonance imaging (MRI) method is presented for quantitatively mapping the physical response of a material to harmonic mechanical excitation. The resulting images allow calculation of regional mechanical properties. Measurements of shear modulus obtained with the MRI technique in gel materials correlate with independent measurements of static shear modulus. The results indicate that displacement patterns corresponding to cyclic displacements smaller than 200 nanometers can be measured. The findings suggest the feasibility of a medical imaging technique for delineating elasticity and other mechanical properties of tissue.

The spatial and temporal pattern of strain created within solid objects subjected to dynamic mechanical stress is of great interest in many disciplines of science and engineering. For instance, the dynamic strain distribution associated with propagation of shear and compression waves is of substantial importance in materials science and in ultrasonics, where such information may be used to predict dynamic behavior and demonstrate structural integrity and composition. However, there have been few noninvasive methods for measuring the three-dimensional strain effects (or strain dyadic) created by mechanical waves within materials (*1*). We describe a method to spatially map and quantitate displacement patterns corresponding to harmonic mechanical waves with amplitudes of 1 μm or less in tissue and other materials with a nuclear magnetic resonance (NMR) technique. Strain and other mechanical characteristics related to wave propagation can be computed from these displacement patterns. Such images could be used to noninvasively deduce important mechanical properties of the internal structure of a wide range of solid materials.

In the presence of a magnetic-field gradient, the motion of nuclear spins causes a phase shift ϕ in an NMR signal (*2*) given by

$$\phi = \gamma \int_{0}^{\tau} \mathbf{G}_{\mathrm{r}}(t)\cdot\mathbf{r}(t)dt \tag{1}$$

where γ is the gyromagnetic ratio characteristic of the nuclei, τ is the time duration of the gradients after excitation, $\mathbf{G}_{\mathrm{r}}(t)$ is a temporal function of the magnetic gradient superimposed on the static magnetic field $\mathbf{B}_0$, and $\mathbf{r}(t)$ describes the position of the nuclear spins as a function of time.

If $\mathbf{r}(t)$ represents simple linear motion, then

$$\mathbf{r}(t) = \mathbf{r}_0 + \mathbf{v}_0 t \tag{2}$$

where $\mathbf{r}_0$ and $\mathbf{v}_0$ are, respectively, the position and velocity at time $t = 0$. If the acceleration and higher order terms are negligible, then given an appropriate gradi-

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