Methods), we estimate that the probability of populating unbound translational states upon ν_s(N–H) decay, and hence producing molecular translation, is 6×10^{-5}. Considering both factors (probability of ν_s(N–H) excitation and probability of decay to translation), the small magnitude of the yield measured for pathway M1 is fully consistent with a mode-selective mechanism of inducing molecular translation, which prevails at lower tunnelling currents. When we raise the excitation rate in order to balance the stretch-to-translation decay rate with an additional excitation of the stretch mode, the energy accumulated in the molecule is large enough to activate an additional decay pathway producing molecular desorption, which is mediated by the umbrella mode (N.L. and J.I.P., manuscript in preparation).

Molecular desorption mediated by the δ_s(N–H_3) mode, M2, dominates when we reduce the electron energy to exclude ν(N–H) excitations. This pathway is described by three excitations of the umbrella mode overtones in a ladder-climbing fashion (Fig. 4b). The ammonia gains sufficient vibrational energy to overcome the 600-meV adsorption well. The umbrella mode features a smaller coupling with translational states than that found for the ν_s(N–H) mode. Moreover, our calculations show that δ_s(N–H_3) vibrational states above 360 meV may well populate a long-lived transition state connected with the complete inversion of the molecule, which leads to desorption after an additional excitation. In this process the umbrella mode itself becomes the vibrational state along the reaction coordinate (Fig. 4b), as was previously deduced from ultraviolet and infrared photodesorption experiments[19,20].

It would be interesting to extend this methodology to more complex processes, searching for strategies of controlling and enhancing reactivity at surfaces that may be applied at the macroscopic scale. The controlled environment furnished by the STM allows the detection of reaction mechanisms in the limit of very low yield and very low power irradiation. In such single-molecule studies, we expect that mode-selective strategies will become important in the discovery of reaction pathways that are inaccessible by classical 'thermal' chemistry. □

Methods

To estimate the potential barriers for translation and desorption, we performed total-energy calculations using plane waves and pseudopotentials in the generalized gradient approximation of density functional theory. Using the method of refs 23 and 24, we also estimated chemisorbed ammonia frequencies, the probability of excitation of each mode and their lifetime. The numerical results for the relevant modes of this work are: the symmetric and one antisymmetric stretch mode, ν_s(N–H) and ν_a^1(N–H), at 408 meV and 422 meV (294 meV and 311 meV for ND_3), both with a probability $P_\nu = 6 \times 10^{-4}$ of being excited per impinging electron and a lifetime of 4 ps and 8 ps, respectively; the umbrella mode, δ_s(N–H_3), at 139 meV (104 meV for ND_3), 40×10^{-4} and 25 ps; the scissors modes, δ_a(N–H_3), degenerate at 200 meV (145 meV for ND_3), 2×10^{-4} and 11 ps.

The decay probability, P_m, of a single ν_s(N–H) excitation decaying into the unbound hindered translation levels plus electron–hole excitation is estimated as the fraction $w_{m>30}/w$. $w_{m>30}$ is the sum of damping rates into states m above the barrier for translation, and w is the damping rate of the ν_s(N–H) mode, via excitation of electron–hole pairs and hindered translational states. An upper limit of the translation yield is then $Y_R = P_\nu \times P_m = 3 \times 10^{-8}$ molecules per electron.

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Impact of urbanization and land-use change on climate

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The most important anthropogenic influences on climate are the emission of greenhouse gases[1] and changes in land use, such as urbanization and agriculture[2]. But it has been difficult to separate these two influences because both tend to increase the daily mean surface temperature[3,4]. The impact of urbanization has been estimated by comparing observations in cities with those in surrounding rural areas, but the results differ significantly depending on whether population data[5] or satellite measurements of night light[6–8] are used to classify urban and rural areas[7,8]. Here we use the difference between trends in observed surface temperatures in the continental United States and the corresponding trends in a reconstruction of surface temperatures determined from a reanalysis of global weather over the past 50 years, which is insensitive to surface observations, to estimate the impact of land-use changes on surface warming. Our results suggest that half of the observed decrease in diurnal temperature range is due to urban and other land-use changes. Moreover, our estimate of 0.27 °C mean surface warming per century due to

land-use changes is at least twice as high as previous estimates based on urbanization alone[7,8].

Two methods used in the US to classify meteorological stations into urban and rural to 'correct' the observed surface temperature trends for urbanization effects are based on population data[5] and satellite measurements of night-light[6–8], respectively, and the corresponding estimates of the impact of urbanization differ in magnitude (0.06 and 0.15 °C per century)[7,8]. The finding that atmospheric temperatures as measured by satellites and weather balloons have smaller warming trends than surface observations has been the subject of much discussion[9] centred mostly on the quality of the data, but it could be partially explained by a predominance of land-use effects over greenhouse warming near the surface.

We estimated the impact of urbanization and other land uses on climate change by comparing trends observed by surface stations with surface temperatures derived from the NCEP-NCAR 50-year Reanalysis (NNR)[10]. In the NNR (a statistical combination of 6-hour forecasts and observations), surface observations of temperature, moisture and wind over land are not used[11]. However, atmospheric vertical soundings of wind and temperature (rawinsondes and satellite soundings) strongly influence the NNR, and surface temperatures are estimated from the atmospheric values. As a result, the NNR should not be sensitive to urbanization or land-use effects, although it will show climate changes to the extent that they affect the observations above the surface.

As indicated by Fig. 1 and many other studies, the NNR captures well surface temperature variations caused by atmospheric storms, advection of warm/cold air, and variations in the frequency or track of major storms. In contrast to the actual surface observations, we find no statistically significant difference in the NNR estimation of urban and rural station trends (see Methods). These arguments suggest that we could attribute the differences between monthly or annually averaged surface-temperature trends derived from observations and from the NNR primarily to urbanization and other changes in land use.

We compare the daily maximum and minimum temperatures of 1,982 surface stations located below 500 m in the 48 contiguous United States, and the daily surface maximum and minimum temperatures on a 2.5° gaussian grid from the NNR interpolated to the station locations, both for the period 1950–1999. We compute temperature anomalies with respect to the 50-year mean annual cycle for each site and each data set. Trends are computed as changes in decadal averages in the anomalies to reduce random errors. The NNR (1948 to the present) has been constructed with a model and data assimilation system kept unchanged, but it is affected by changes in the observing systems, especially the introduction of the satellite observing system in 1979. Therefore, in the computation of trends we exclude changes from the decade of the 1970s to the 1980s.

Figure 1 compares time series of 50 years of monthly mean temperature anomalies for Baltimore, a large city in Maryland, including the average decadal difference between observations and the NNR. There is good agreement in the interannual variability, with a correlation of over 0.9, but also a growing trend in the difference between the surface observations and NNR, increasing to 1.4 °C during the 1990s, a difference we attribute to urbanization and other surface changes that do not affect the NNR. A similar analysis on all surface stations (Supplementary Fig. 1) indicates a 50-year correlation of about 0.9 everywhere except in mountainous regions, where it is between 0.4 and 0.7, which is why we only include stations located below 500 m. The correlation is also lower in the west coast, possibly owing to the proximity of mountains or to low data density in the Pacific Ocean. Decadal trends can be locally dominated by interannual and decadal variability of the temperature due to anomalies in the circulation rather than to land use change—effects that are excluded by taking the differences between surface and NNR temperatures.

The decadal trend averaged over the two separate 20-year periods (1980–1999, and 1960–1979) is computed for every station and averaged in boxes of 0.5° latitude by 0.5° longitude, with an overall average computed over all the boxes. Figures 2 and 3 show the average trends for the observations and from the NNR, and also the difference between these two trends, which is at least partially attributable to changes in use of the land surface.

The maximum temperature (Fig. 2) shows a warming trend in the observations in the eastern and western US and a cooling trend in the Midwest, with a slightly negative overall average of −0.017 °C per decade. The NNR is similar but smoother, with an average of +0.008 °C per decade. The difference between the observed and NNR trends is somewhat negative in most of the country east of the Rockies, but is strongly positive in California and to a lesser extent, in Oregon and Washington, with an average difference of −0.025 °C per decade.

The minimum temperature (Fig. 3) observations show a much

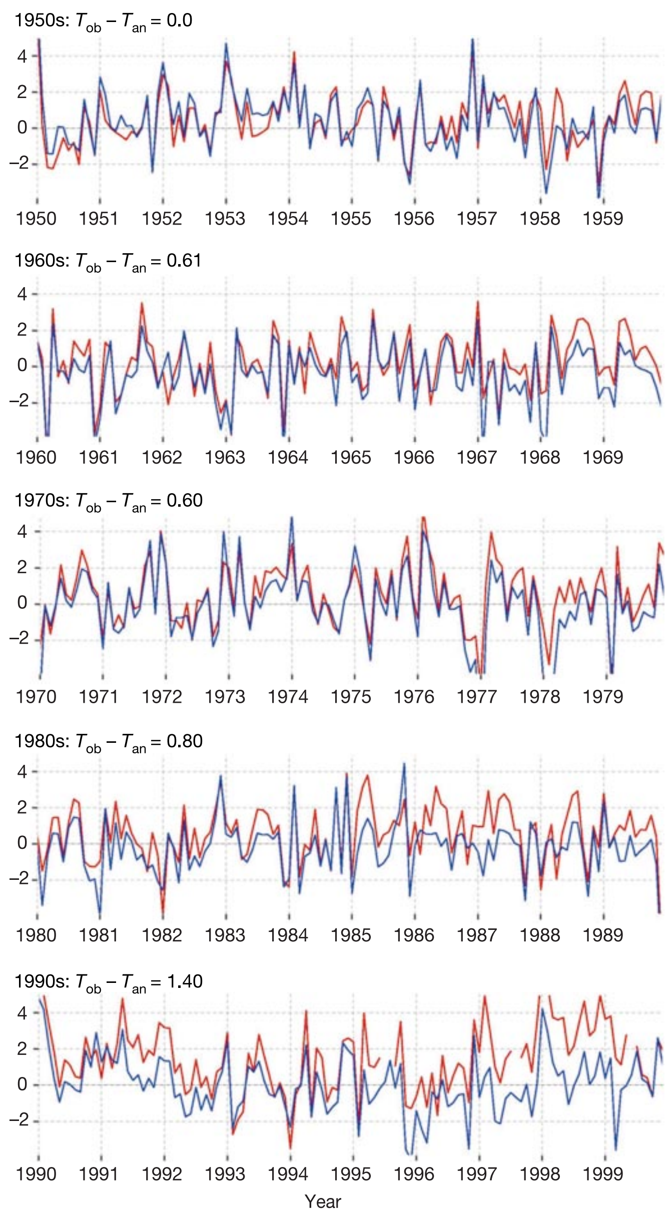

Figure 1 Comparison of monthly mean station and NNR surface temperature anomalies with respect to their annual cycles for the city of Baltimore, Maryland, USA. T_{ob}, observed monthly mean temperature in °C, shown in red. T_{an}, analysed monthly mean temperature in °C, shown in blue. Five decades (1950 to 1999) are shown for comparison.

stronger positive trend in most of the country, with an average of +0.193 °C per decade. In the NNR, the minimum temperature increases everywhere except in the Midwest and California, with an average of +0.113 °C per decade. The difference in minimum temperature trends between observed and NNR values is positive in most of the country, especially in California, with an average of 0.080 °C per decade (40% of the observed trend).

Supplementary Fig. 2 shows the trend in the difference between maximum and minimum temperatures or diurnal temperature range (DTR). In the observations the DTR trend is strongly negative in most of the country with an average decrease of −0.210 °C per decade. The NNR also shows a general decrease of DTR, with a national average of −0.105 °C per decade so that about half of the decrease in DTR could be attributable to surface changes.

The daily mean temperature observation trends (Supplementary Fig. 3) obtained as the average of the maximum and minimum temperatures show an increase in most of the country, with an average trend of +0.088 °C per decade. The NNR trends have an average of +0.061 °C per decade. Of the two "urban correction" estimates[7,8], our estimate of 0.27 °C per century attributable to land use is closer to the estimate based on the night-light urban effect (see centre bottom of plate 3C in ref. 7) than to the estimate based on population density (see centre bottom of plate 3B in ref. 7). It should be noted that our observed daily mean temperature trends (Supplementary Fig. 3a) are different from previous 50-year or 100-year trend estimates (see plates 3A and 7A in ref. 7) because in our computations we did not include (1) the decadal trends corresponding to the 1980s–1970s and especially the 1960s–1950s, and (2) urban and non-urban data adjustments. The non-urban adjustments tend to be strongly positive except over the Rockies (see plate 3B in ref. 7), so that if we had added them to the raw observations, our estimate of the land-use impact on the mean temperature trends would have been geographically similar but larger.

Although it is not possible definitively to attribute the differences between the observation and the NNR temperature trends solely to land use, including urbanization, agriculture and irrigation, our results are compatible with such an interpretation. The well-known 'urban heat island' effect actually takes place at night, when buildings and streets release the solar heating absorbed during the day. At the time of the maximum temperature the urban effect is one of slight cooling, owing to shading, aerosols, and to thermal inertia differences between city and country that are not currently well understood[12].

The effect of agricultural development, increasing evaporation during the day, would also tend to decrease the maximum temperature: irrigation would increase the heat capacity of the soil, thus increasing the minimum temperature. Therefore, both urbanization and agriculture effects could be consistent with the general increase in the minimum temperature and slight decrease in the maximum temperature, and contribute to the reduction in the diurnal temperature range shown in our estimates east of the Rockies (Figs 2c and 3c).

This implies that the comparison of urban and rural stations without including agricultural effects would underestimate the total

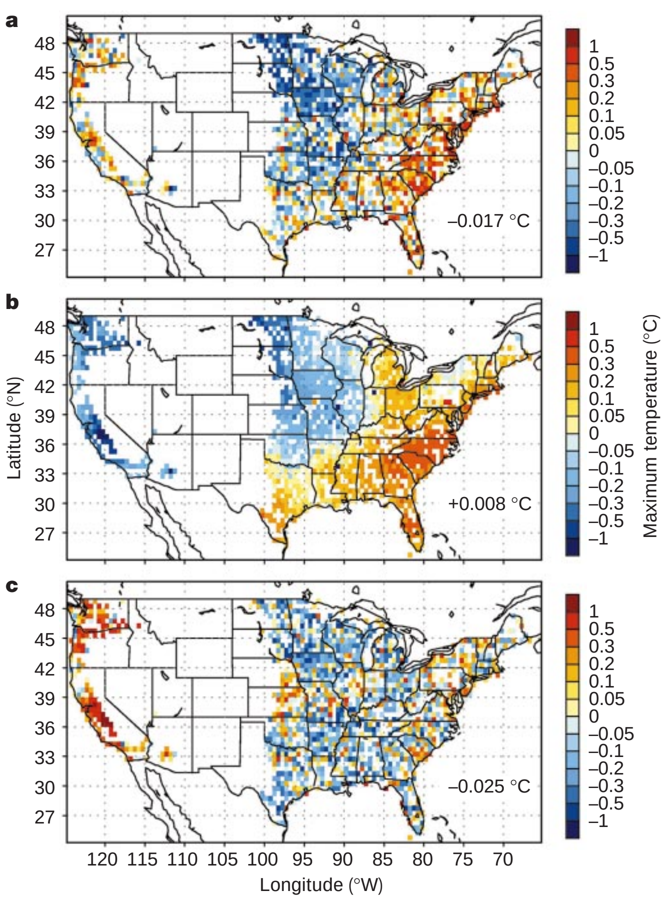

Figure 2 Decadal trend of the maximum temperature averaged for every US station below an elevation of 500 m. Each value (in °C per decade) was calculated from the average of the '1990s minus the 1980s' and the '1970s minus the 1960s' maximum temperatures. The station values are displayed as averages in boxes of 0.5° latitude by 0.5° longitude. Blank boxes indicate that none of the 1982 stations is within the boxes, and the national average is the average of these boxes. The average value of the trend is indicated in each panel. **a**, Station (observed) maximum temperature trends. **b**, NNR (analysed) maximum temperature trends. **c**, 'Observed minus analysed' maximum temperature trends.

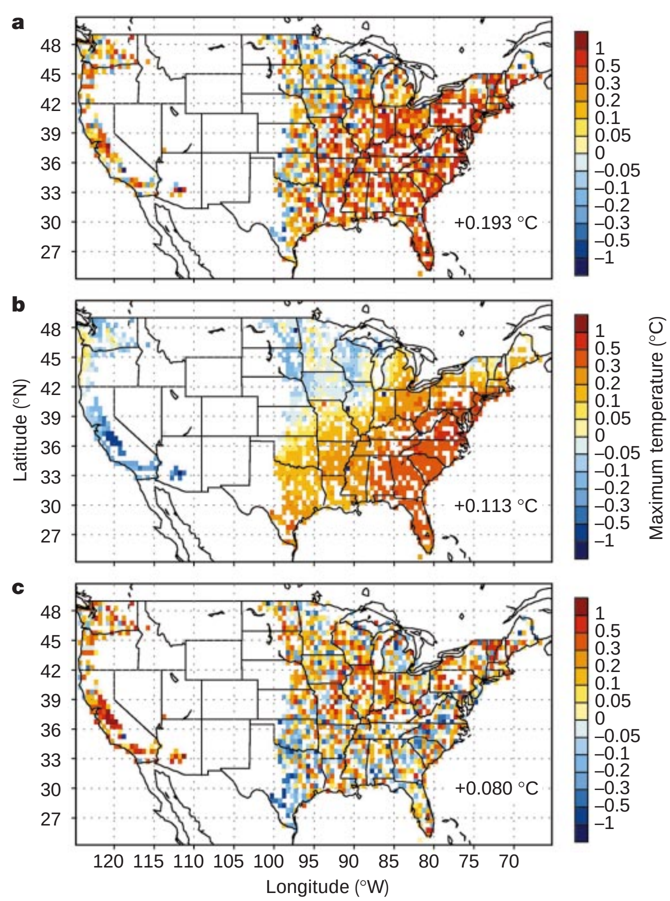

Figure 3 Decadal trend of the minimum temperature averaged for every US station below 500 m. Legend as for Fig. 2 but for the minimum temperatures.

impact of land-use changes. More studies are needed, including a comparison of geographical distribution of NNR trends with other upper-air observations, such as rawinsondes and satellites, a more precise definition of the urban and rural observing stations, and the impact of other human activities such as contrails and aerosols that can also reduce the diurnal temperature range[13].

Our method can incorporate updated observations as they become available, can be applied to land stations throughout the world, to other variables such as humidity and winds, detect seasonal trends, and signal changes in station locations that are otherwise difficult to identify. □

Methods

Data

For the surface observations, we use the daily surface maximum and minimum uncorrected surface station temperatures from the National Climate Data Center (NCDC) 'Cooperative Summary of the Day' data set over the 48 contiguous states of the United States for 1950–1999. For the NNR, we use the global daily surface maximum and minimum temperatures gridded on 2.5° gaussian boxes, also for the period 1950–1999.

Analysis

We interpolate linearly the gridded NNR data to each observational site, and only consider the sites that have a total of at least 480 (whole) months of observations. In addition, because the NNR has surface heights different from those of the real locations, and extrapolations underground can introduce errors overwhelming the signal of the real trends (Supplementary Fig. 2), in the computation of the trends we only consider sites with elevations lower than 500 m. There are 1,982 US surface stations satisfying these two conditions. We obtain monthly means by averaging daily data; daily mean temperatures are obtained by averaging maximum and minimum temperatures, and daily temperature ranges by subtracting the minimum from the maximum temperature.

Because the NNR can have systematic differences with observations, especially near the surface, owing to deficiencies in the model forecast or the method of assimilation, we remove the 50-year monthly mean annual cycle for each site from both the observations and the NNR. We are thus comparing anomalies with respect to the 50-year mean annual cycle. In the results we present both comparisons of the 50-year time series and trends. The trends are computed as changes in decadal averages in order to reduce random errors. We only consider two decadal trends: the decade 1990–1999 minus 1980–1989, and 1970–1979 minus 1960–1969. We do not include in the trends the difference between the decades 1960–1969 and 1950–1959, because the observing system during the 1950s was considerably less reliable than in later decades, and it underwent significant scheduling changes during 1958 (ref. 11).

In addition, we have to address changes in the observing systems, especially the introduction of the satellite observing system (of which the most important is the TIROS-N Operational Vertical Sounder, TOVS) starting in 1979. These two major changes are the main reason why trends in the NNR need to be carefully estimated. We therefore do not include the changes 1980–1989 minus 1970–1979. The two decadal changes that we keep correspond to the 1990s minus 1980s (20 years with satellite data), and 1970s minus 1960s (20 years essentially without satellite data). Thus, when we average them we obtain decadal trends from two independent and largely homogeneous 20-year periods.

We compared the 1990s versus 1980s trend of 775 stations classified as urban versus 167 stations classified as rural. The mean surface temperature increased by 0.31 °C for the urban stations and 0.13 °C for the rural stations, with standard deviations of about 0.5 °C each. The difference between urban and rural warming, 0.18 °C, is significant at a 99% level of significance. The trends for the reanalysis station estimates are 0.26 °C for urban and 0.25 °C for rural, with standard deviations of about 0.22 °C, and the difference 0.01 °C between urban and rural is insignificant, showing that the NNR is insensitive to surface effects.

In the time series we compute the 1950–1959 average temperature difference between the NNR and the surface station at each station and subtract it from the NNR. This forces the two time series to have the same 10-year time average during the 1950s and is done for display but does not affect the computation of the trends or correlations.

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Field sports and conservation in the United Kingdom

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Many natural habitats exist on privately owned land outside protected areas[1], but few governments can afford to enforce or subsidize conservation of this biodiversity. Even in some developed countries, conservation subsidy schemes have only achieved limited success[2–4]. Fortunately, some landowners may be willing to accept management costs in return for other benefits[5], although this remains controversial when it involves the killing of charismatic species. For example, participants in British field sports, such as fox hunting and game-bird shooting, may voluntarily conserve important habitats that are required by quarry species[6–8]. Here we report results from a multidisciplinary study that addressed this issue by focusing on three sites across central England. We found that landowners participating in field sports maintained the most established woodland and planted more new woodland and hedgerows than those who did not, despite the equal availability of subsidies. Therefore, voluntary habitat management appears to be important for biodiversity conservation in Britain. Current debates on the future of field sports in Britain, and similar activities globally, may benefit from considering their utility as incentives to conserve additional habitat on private land.

Private landowners play an increasingly important role in biodiversity conservation[1]. This is especially important where habitats form isolated remnants in an agricultural matrix, and it is politically difficult to establish large protected areas[9]. This is typified by the situation in Britain, where farmland covers 76% of the country and increases in agricultural efficiency have caused great declines in biodiversity[7,10,11]. The British government has responded by introducing legislation to protect important habitats and species on public and private land[12–14], as well as establishing subsidy schemes[11,15]. However, conservation legislation remains unpopular