The Dependence of Urban Scale Oxidant Chemistry on Boundary-Layer Processes and Regional Scale Transport

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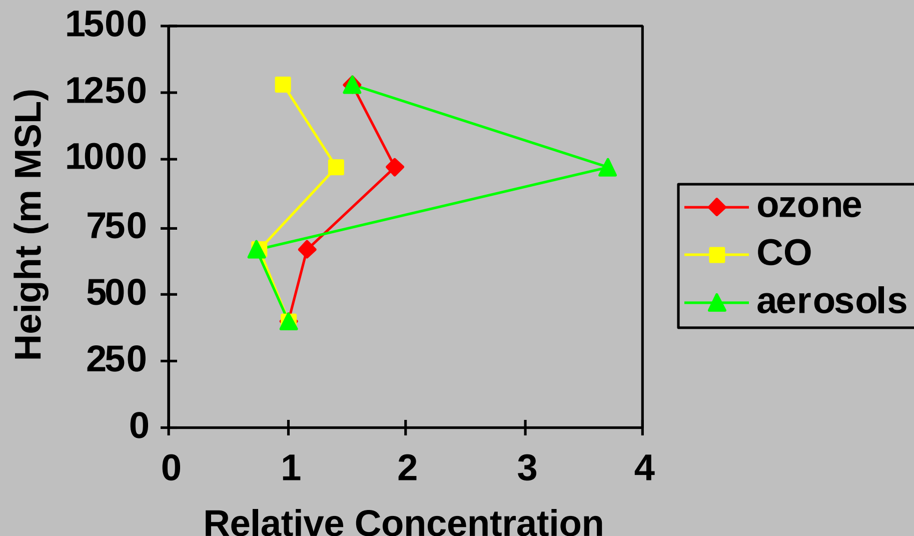
The problem: The downward mixing of layers of pollutants into the boundary layer and the transport of polluted air from upwind sources are thought to play important roles in high ozone episodes. However, the processes through which elevated polluted layers form, are transported, and mixed vertically are not well understood.



Example of measured pollutant profiles over Connecticut. The formation of these layers is thought to be related to the cyclic pattern of emissions in the evolving urban boundary layer and the boundary layer's interaction with the free troposphere.

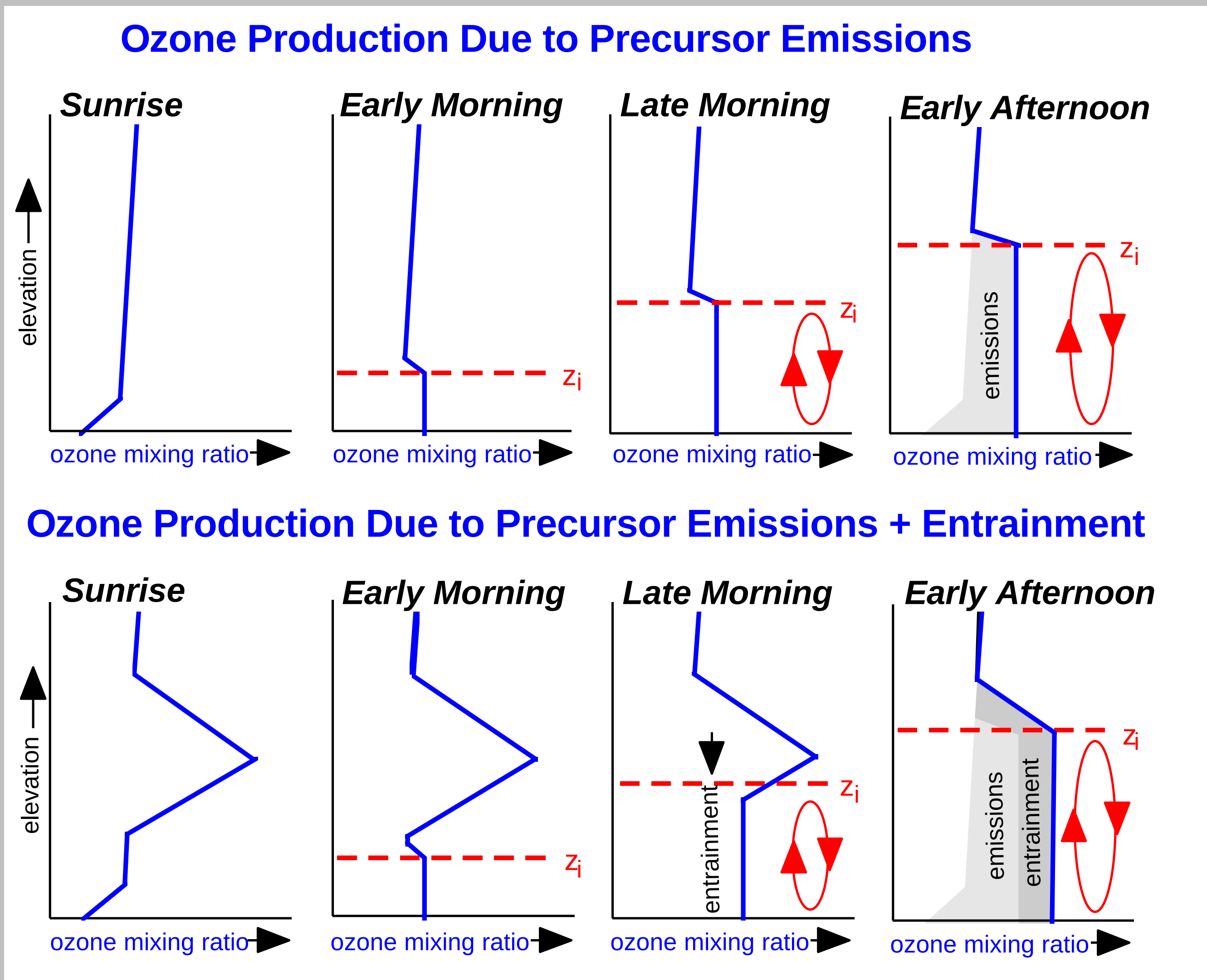
Questions:

How do near-surface concentrations of ozone depend on

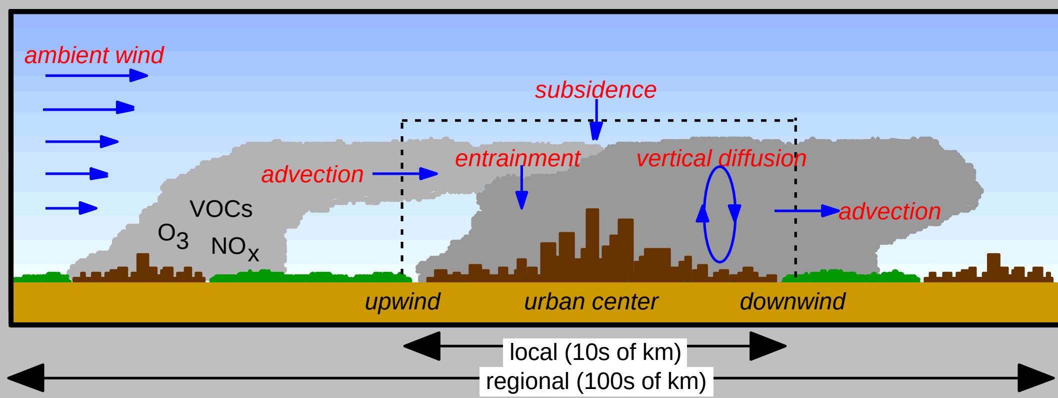
- **the vertical distribution of ozone and its precursors,**
- **the evolving boundary-layer structure of the atmosphere, and**
- **the vertical transport and mixing of material between surface and elevated layers?**

What are the relative contributions to the local vertical distributions of ozone, VOCs, and NO_x from

- **local sources and**
- **the advection of material from upwind regional sources?**



Conceptual diagram depicting the development of ozone profiles during convective boundary layer growth as a result of (top) surface emissions and (bottom) surface emissions plus entrainment.



Schematic diagram illustrating some of the local and regional boundary-layer transport and mixing processes that can affect local air chemistry.

Approach:

Measurements:

- **aircraft (O_3, NO_y, CO, hydrocarbons, winds, temperature, humidity)**
- **radar wind profilers (winds)**
- **airsondes (temperature, humidity)**

Modeling:

- **nested mesoscale model (RAMS) with 4 dimensional data assimilation**
- **Lagrangian particle dispersion model**
- **coupled RAMS/ limited-area GChM photochemical model**

Siting strategy:

- **Initial study in an isolated urban area with relatively small influences from major upwind industrial or urban sources (e.g. Phoenix)**
- **Progress to sites with more complex local and regional upwind sources (e.g., Nashville, northeastern U.S.)**

Specific questions for Phoenix study

Observational:

- **What is the relative importance of recirculated polluted air to ozone and aerosol concentrations in Phoenix?**
- **What are the time scales of such circulations (same day, multi-day)?**
- **What is the importance of thermally-driven wind systems for the transport of isoprene, olefins, and other hydrocarbons into Phoenix from the surrounding mountains?**
- **Are external pollutant sources a significant source of ozone for Phoenix?**

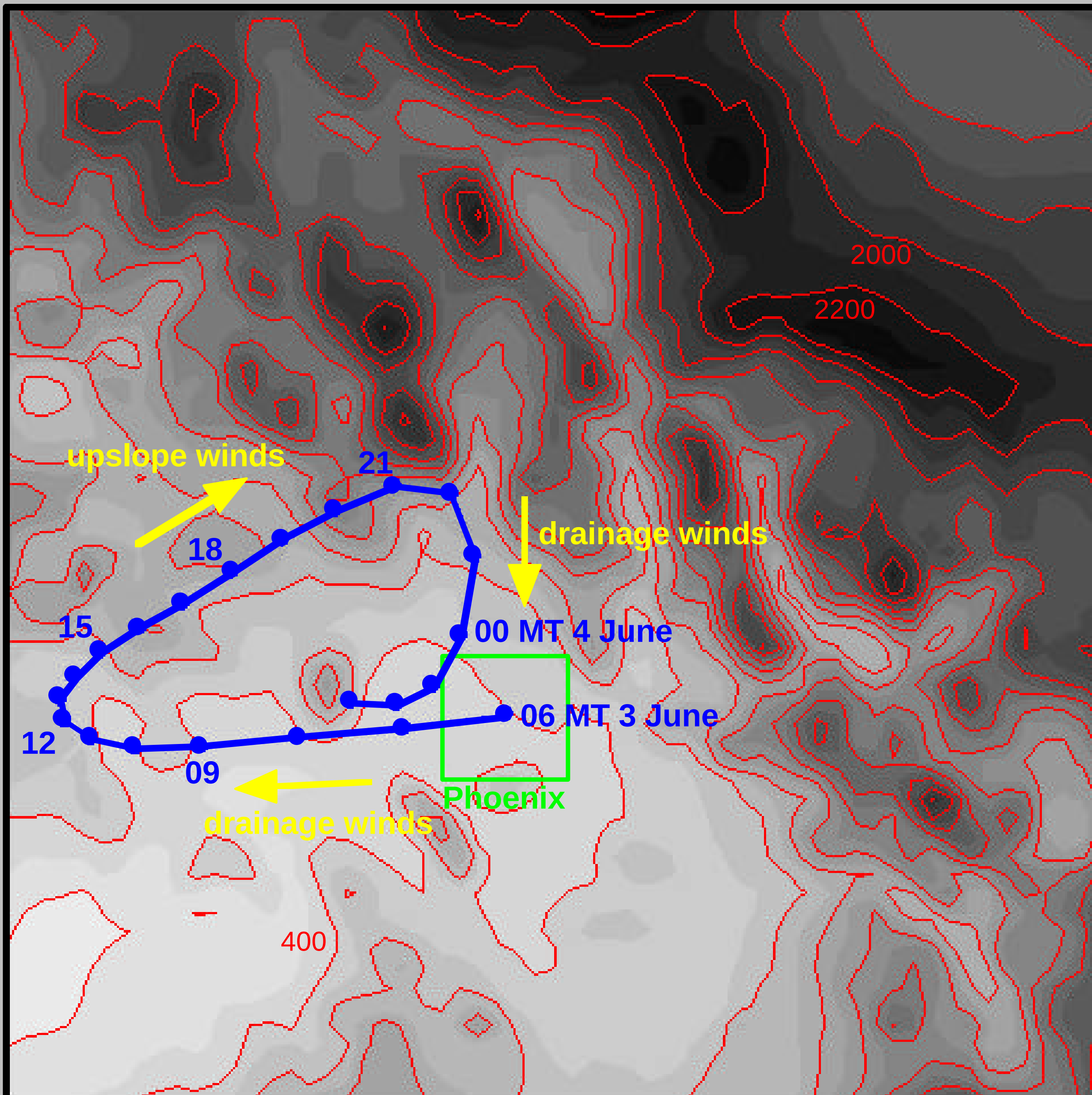
Specific questions (continued)

Modeling/observational:

- **Is the production of OH by olefins significant compared to that produced from the O(1D) reaction with water?**
- **How do the near-surface concentrations of ozone depend on the initial vertical layering of NOx, HC, O_3, etc.?**
- **What is the relationship, if any, among mixed layer depths, intensity of recirculations, and surface ozone concentrations?**

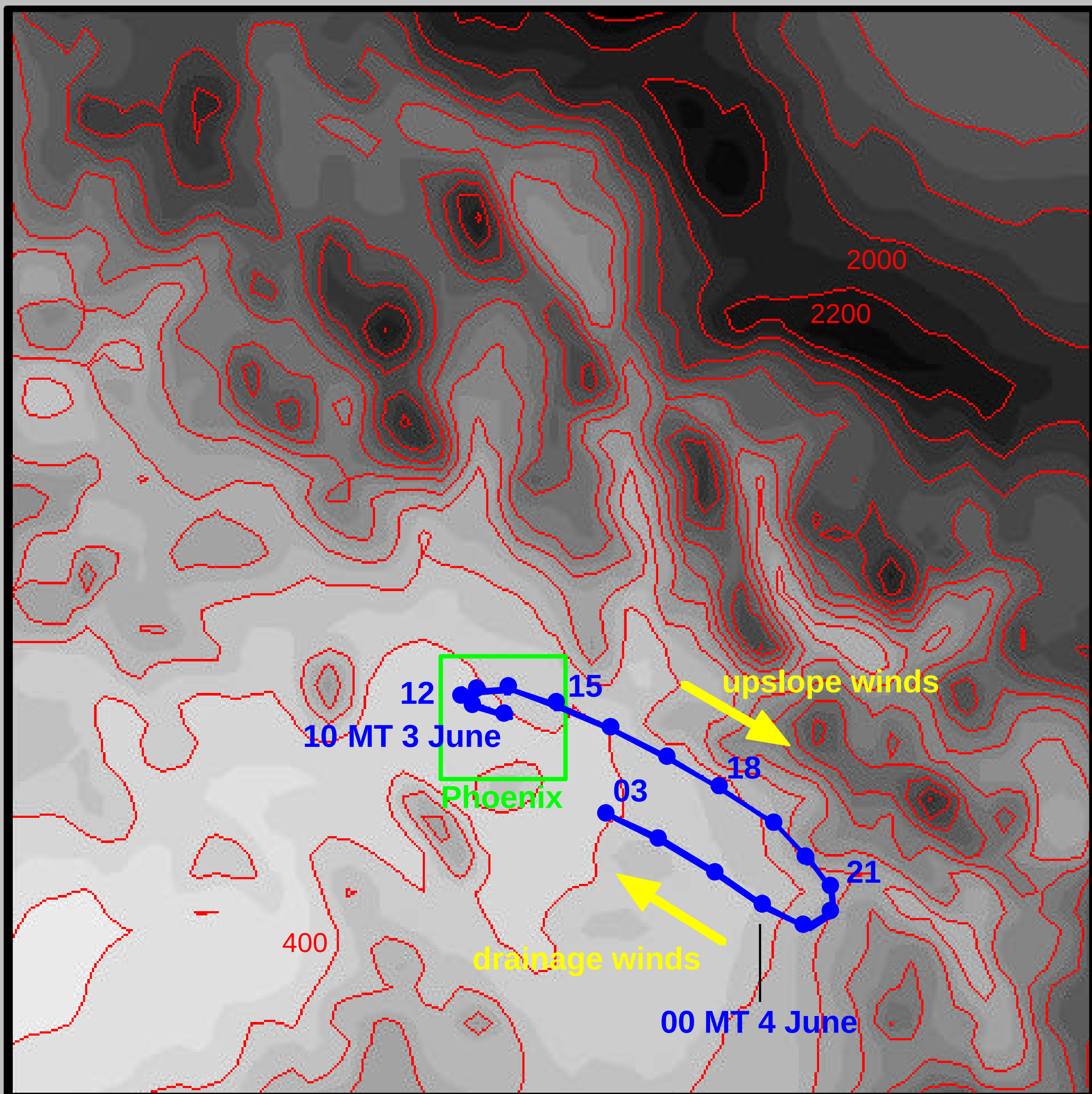
The following three figures show simulated trajectories of tracer particles released near the surface of Phoenix during the period 3-4 June 1996. The simulations were done with the RAMS mesoscale model operating in a nested grid configuration. The innermost domain shown here is approximately 300 km on a side with a horizontal resolution of 5 km. The model was initialized with the analysis fields from the ETA model and run for 48 hours. During 3-4 June a high pressure system was located over the western U.S. and featured light winds aloft - conditions favorable for the development of thermally driven flows in the Phoenix area.

The figures show the paths of particles released at 0600, 1000, and 1400 Local Standard Time. The influence of upslope and downslope flows is clearly evident and results in significant recirculations of material released during the morning hours. Turbulent diffusion was turned off for the trajectory calculations; a simulation carried out with turbulence switched on shows considerable spreading of the particle "plume".



• Material emitted early in the morning is transported to the west before being advected up the slopes of the mountains.

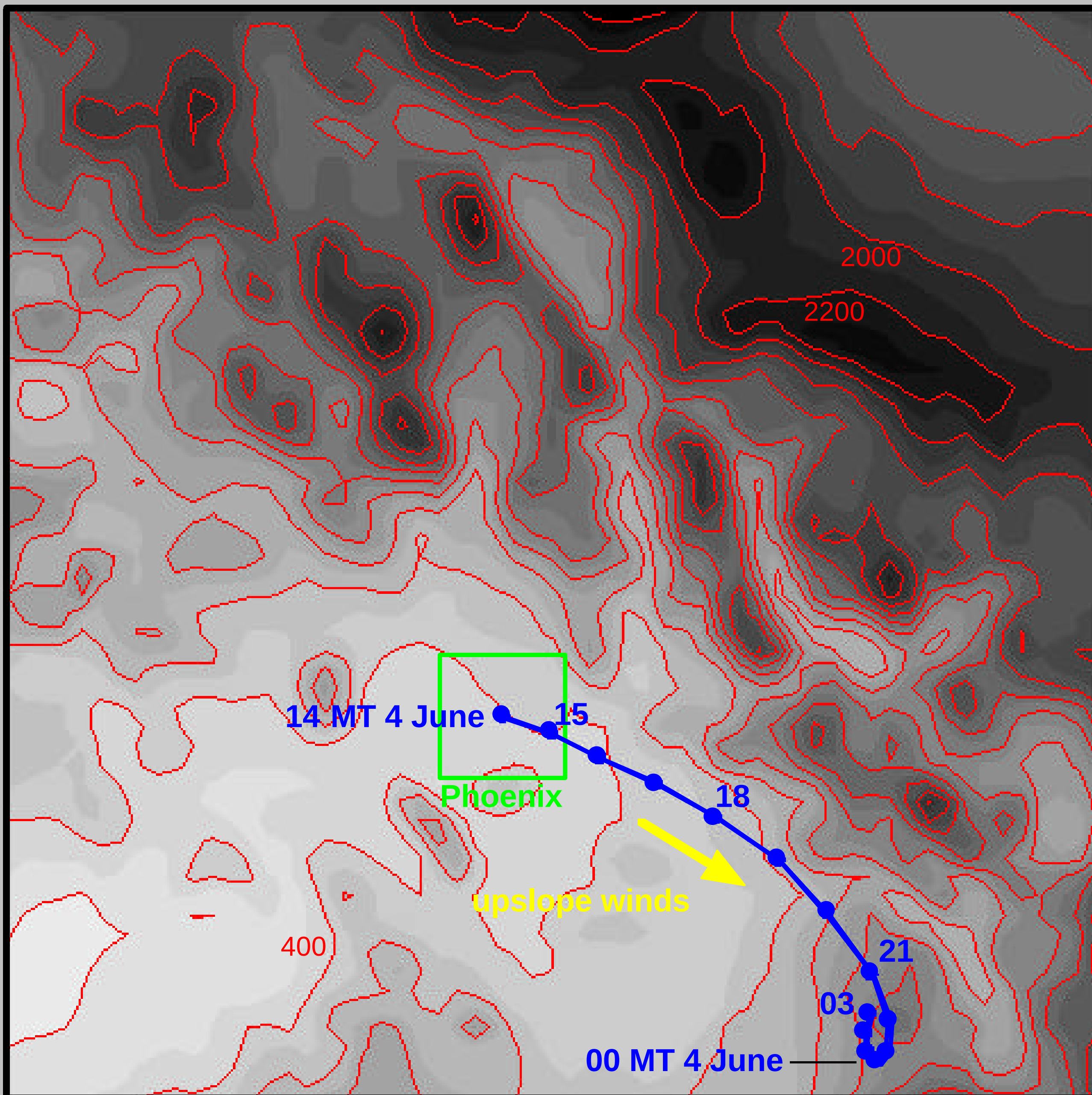
• Drainage winds may advect previously emitted pollutants back over Phoenix during the evening.



• **Material emitted later in the morning is transported to the mountains to the east and southeast.**

• **Light winds around noon produce nearly stagnant conditions over Phoenix.**

• **Drainage winds may advect previously emitted pollutants back over Phoenix during the early morning.**



- Material emitted during the afternoon is transported toward the mountains to the southeast.

- These pollutants are not advected back toward Phoenix.