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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The impact of free convection on late morning ozone decreases on an Alpine foreland mountain summit" *by* J.-C. Mayer et al.

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The authors have presented a very interesting analysis of significant depletions in mountain top ozone during the morning, resulting from free convection of boundary layer air. And while there is good evidence pieced together from different instrumentation at three different observing sites, supporting the free convection idea, there are a number of questions that the paper fails to address.

The authors first describe a *typical event* from the observed events during the SALSA campaign: mean depletion was 52%, mean initial O_3 was 45.5ppb, mean minimum O_3 was 22 ppb, mean duration was 42 minutes. However, they focus on data that

was actually most extreme: September 5, recorded an 81% depletion from 41ppb to a minimum of 7ppb, and lasted for 118 minutes, or almost two hours. One of the most seemingly relevant aspects of this event was the NO concentration, which spikes from what must be a few ppt to 12ppb, and then decays away. This begs the questions, was there a corresponding extreme increase in NO for each of these events? If so, that might be as relevant overall as the O_3 observations. There is also the question of whether in-situ chemical destruction of ozone is occurring as a result of the pulse of boundary layer air. It is certainly the case that the boundary layer ozone at the BASE site was fluctuating between 10 and 20 ppb, and yet, this extreme event at the TOP site resulted in a drop from 41ppb to 7ppb. In their discussion, the authors have implied that the processes which could lead to these O_3 drop events are independent, and yet it seems fairly obvious that the injection of air up to the elevation of the TOP site, via free convection of air near the foot of the mountain will in fact result in a transient change of airmass, and more importantly, it could result in a temporally limited O_3 sink at the mountain summit, a chemical sink. Chemistry and dynamics are not mutually exclusive.

The authors make the point that these free convection events were buoyancy driven by bubbles of warm, moist air rising during a period of light wind. Was there a corresponding increase in the water vapor mixing ratio observed at the TOP? This is implied by the increase in specific humidity as shown by the time height cross section of tether-sonde data from the BASE site. There should have been a corresponding increase in the RH, or a change in theta-e as measured at the TOP. Was this observed?

It would seem that some of these things might have been analyzed during SALSA if indeed it was investigating OH contributions from nitrous acid. It would appear very consistent that HONO, a nocturnal reservoir species that is also soluble, could be released in warming, lower level surface air that is evaporating fog, or mist, possibly even dew, then photolyzed to OH and NO. This brings another question to mind, and the authors should have data to address it. Was the base site saturated earlier in the morning

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on all of these event days (RH=100% or almost 100%?). In fact, is that a significant part of these free convection events, the evaporative heat added to the lower boundary layer results in the subsequent destabilization? The TOP site shows the clear condition, and global radiation curve for the September 5 event in figure 4. But have you explored the use of the Bowen Ratio for the base site for this time period, it would represent the ratio of the energy flux upward as sensible heat to the energy flux used for evaporation? Given the timing, it seems very plausible that you are burning off a mist or light fog, or dew in the vicinity of the lower forested site. Consider the sun rises, and some surface heating begins, but before the sensible heating increases significantly to build the convective boundary layer at the BASE sites, there are pockets of free convection (driven by buoyancy from the evaporative energy flux overcoming shear in the light wind) which produce eddies or large bubbles of air with near-surface chemical concentrations abrubtly injected aloft. I think it would be helpful to discuss this by considering more data than just ozone. I recommend you add both NO data, and RH to Table 2 for the campaign events, these gases should also be changing. It would also strengthen your argument if you add something other than just wind and T, eg., add virtual temperature, equivalent potential temperature, or RH to figures 3 and 4.

The authors have noted that the magnitude of these events vary temporally, is it possible to estimate the magnitude (size) of the freely convected eddy based on the duration of the depletion event and the local wind speed?

Finally, at the outset of this paper, the authors suggest this phenomenon should somehow be removed (filtered) from the data so as to not affect derived statistical descriptions of ozone. Quote: Furthermore, if such events occur frequently, it must be of considerable concern for all measurements, being made at comparable sites, as such a high frequency of occurrence could, if not filtered, affect derived statistical descriptions of the 20 mean temporal behaviour of the trace gas. End quote

However, considering the fact that these events occur frequently, and are likely to occur at comparable sites, it would seem that this kind of data manipulation, i.e. filtering,

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would be unwise. The point is, long term observations, effectively monitoring data, should *categorically not* be filtered to remove specific natural events that cause fluctuations in the data (remember the ozone hole!). Scientific investigations, indeed, like this paper, that attempt to understand why significant data fluctuations occur, and try to determine the recurrence of particular types of events are in fact what is needed. More thoughtful, detailed investigations like this one that recognizes the interplay between chemistry and meteorology through marshalling a variety of different observations are exactly the kind of integrated analyses we need. But please, *do not* throw out the data!

Specific grammatical errors or suggestions for the text:

Page 1. Rewording: Futhermore, if such events occur frequently, it warrants further study, particularly for measurements being made at comparable sites. Because high frequency occurrence may affect the statistical description of the mean temporal behavior, these events should be well understood.

General terminology: Rather than refer to noticeable events, eye-catching drop, or O-3 drop, I suggest you refer to them as significant O_3 depletion, or perhaps significant negative excursions in O_3.

Page 6. However, we explicitly use the buoyancy flux..in the morning hours, where we will focus. Change to: ..in the morning hours, the period of focus.

A more than 5 years long monitoring dataset;change to: A dataset extending over 5 years.

This was done to get evidence whether the Salsa field observations; change to: This was done to determine if the Salsa field observations.

Page 9: To get a closer insight into the processes being possibly active in the ABL at these days..change to: To get better insight into the potential processes active in the ABL on these days

Page 17. As the air close to the ground was still pooer of O_3 but rich in CO_2 and H_20,

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a small layer with lower O_3 and higher CO_2 .. What about the NO levels?

Page 18. Do you know that the drop is air temperature mentioned in point 1 is not the result of evaporative cooling from burning off of fog or mist? I think you should discuss this.

Page 20. Futhermore these numbers has to be taken as a minimum estimation, as the convectively lifted air masses have not necessarily have to be advacted towards the TOP site or has to be trapped just at the right altitude to hit the TOP station. Change to:

Furthermore, these numbers have to be taken as a minimum estimation, as the convectively lifted air masses do not necessarily have to be advected towards the TOP site, and they do not have to be trapped at just the right altitude to be intercepted by the TOP station.

At 18 percent of the days..change to: On 18 percent of the days.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 5437, 2008.