4.9. Air pollution in Switzerland since the early 1990s

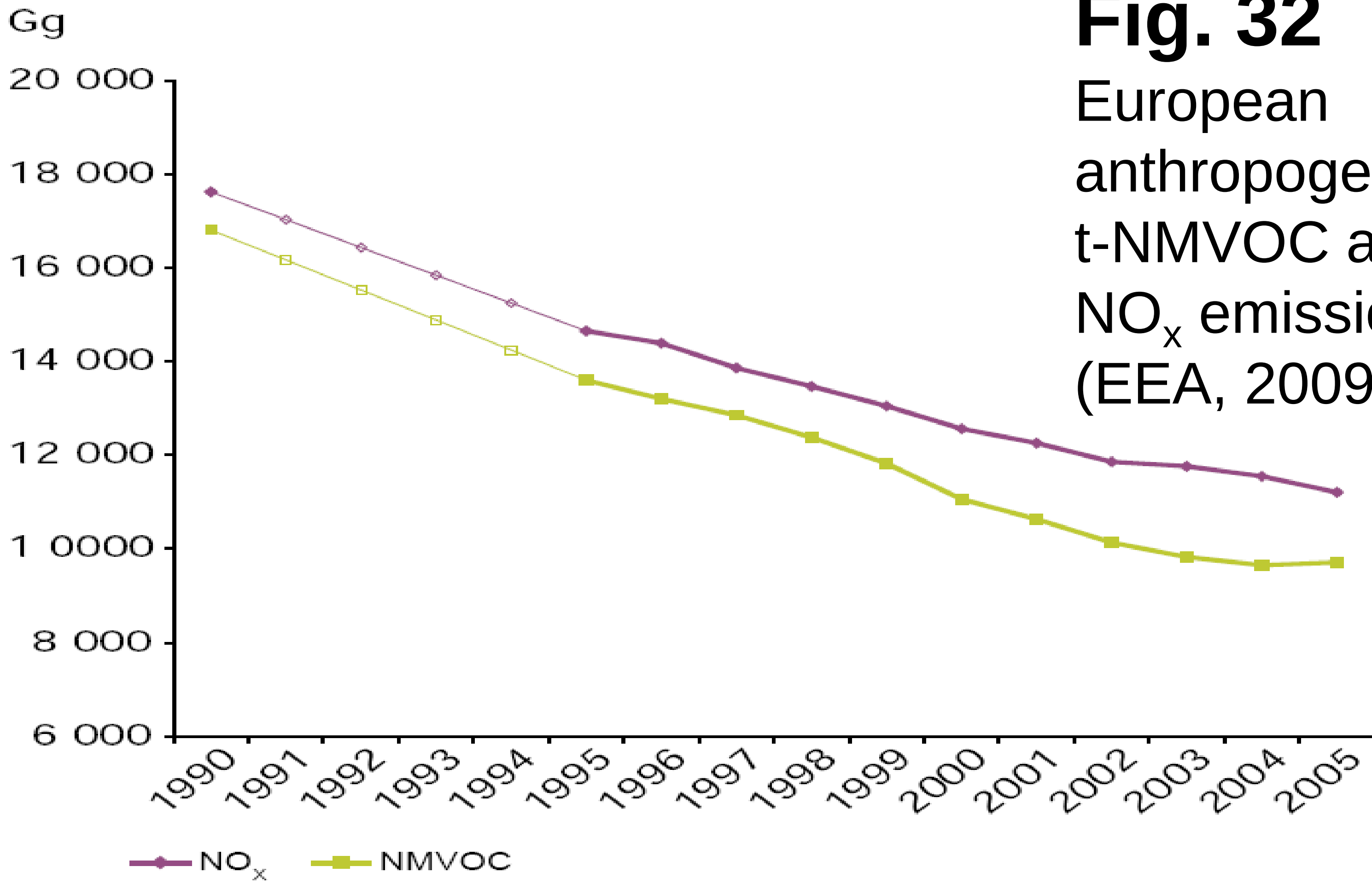


Fig. 32

European anthropogenic t-NMVOC and NO_x emissions (EEA, 2009)

Fig. 33: Total national emissions of NO_x (as ktonnes NO_2) for 1990, 1995, 2000 and 2005 for seven selected countries (from EEA, 2009)

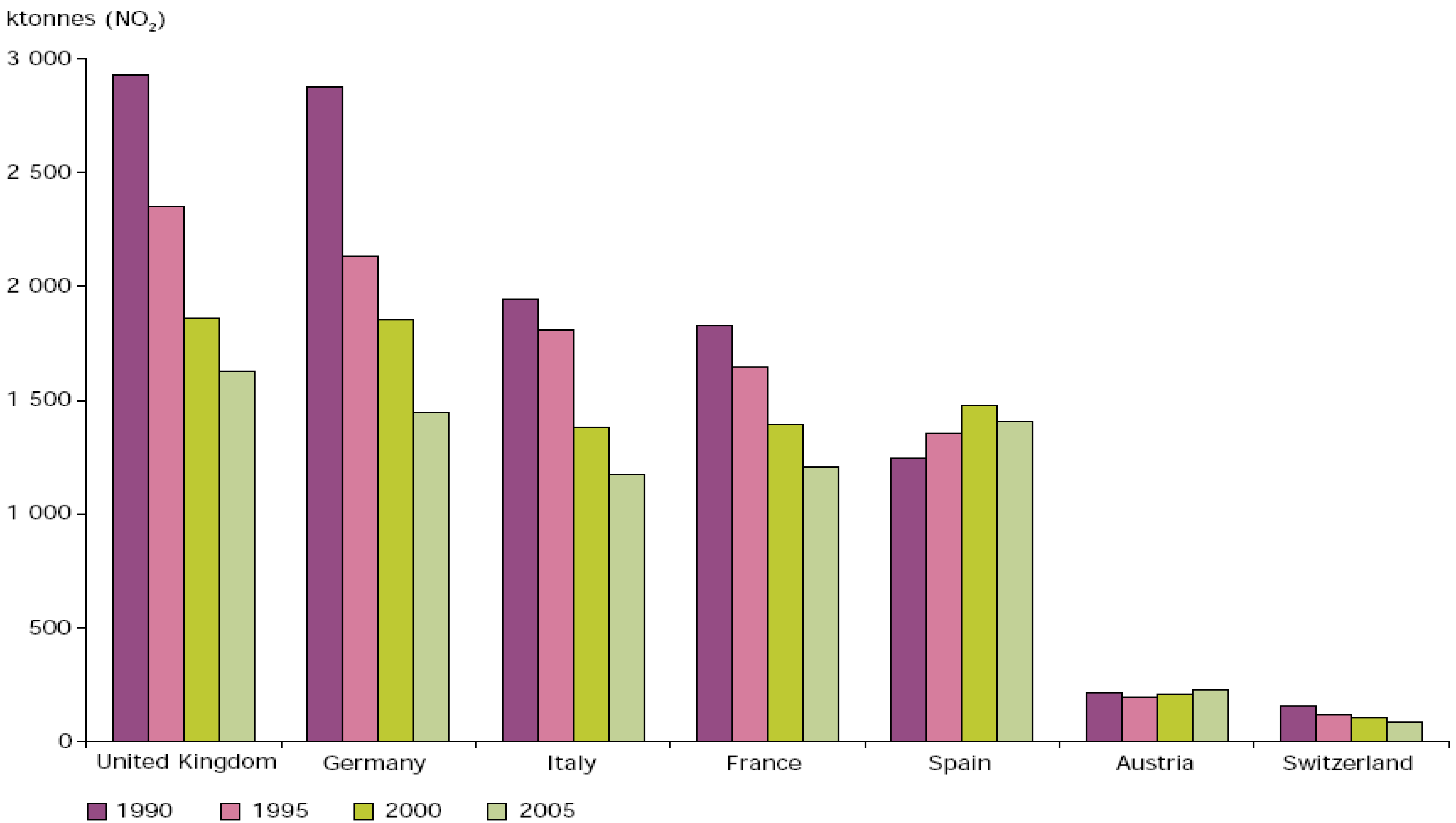
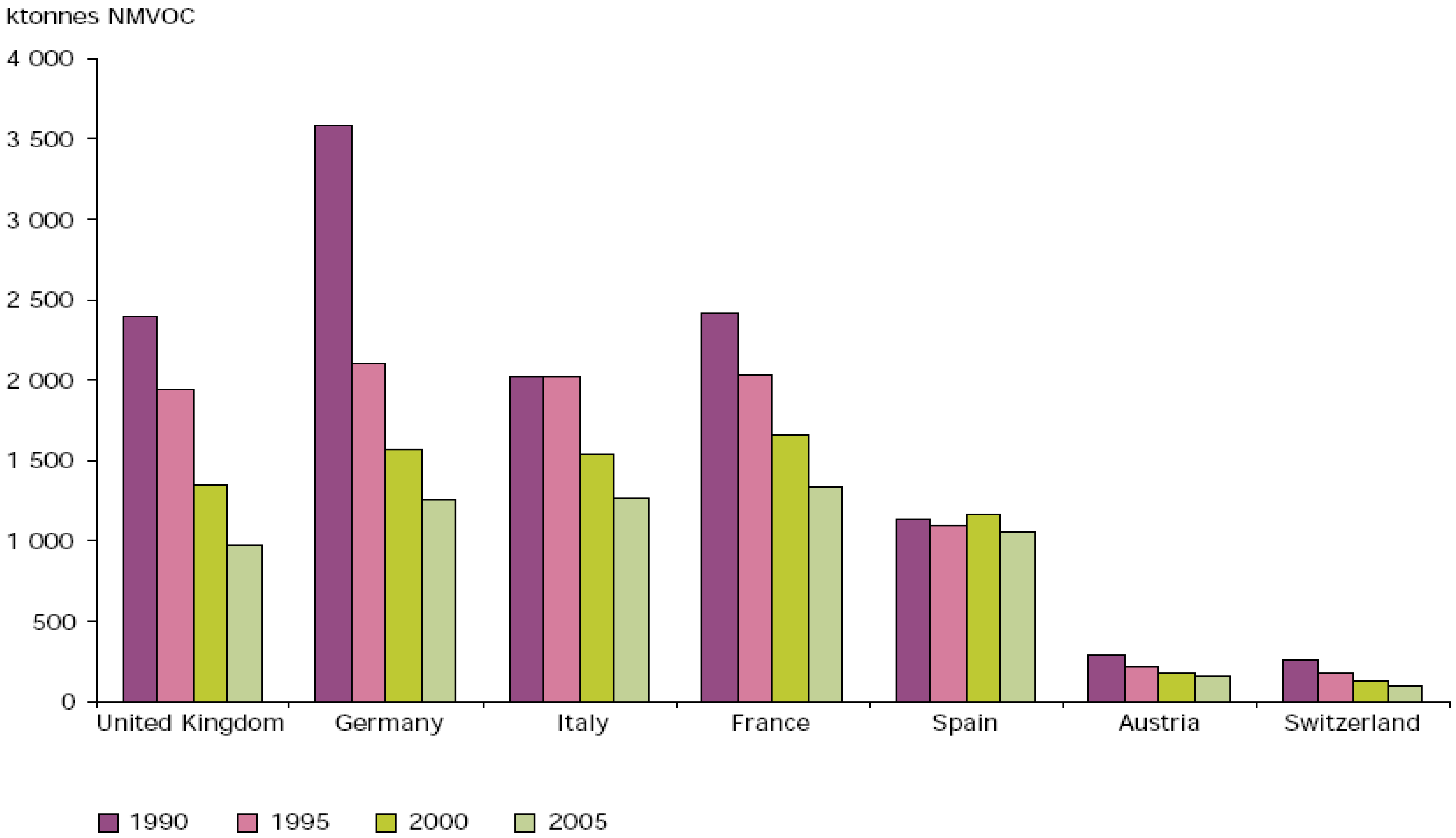


Fig. 34: Total national emissions of t-NMVOCs (in ktonnes) for 1990, 1995, 2000 and 2005 for seven selected countries (from EEA, 2009)



Notes to Fig. 32-34: Anthropogenic emission changes in Europe

- Around the end of the 1980s anthropgenic emissions in the industrialized countries started to stabilize and subsequently to decrease because of the introduction of new technolgies introduced to reduce air pollution (comp. Fig. 25 and 26).
- According to the EMEP data anthropogenic emissions in the 1990s strongly decreased in Switzerland and the surrounding European countries because of (new) air pollutant legislation (Fig. 32 - 34).
- Over entire Europe, NO_x and t-NMVOC decreased by approximately 30% between 1990 and 2005 (Fig. 32).
- The amount of decrease substantially varied among the countries: In UK, Germany and Switzerland NO_x and t-NMVOC decreased approximately by a factor of 2 (1990 and 2005), the decreases were smaller in France, and Austria whereas the emission increased in Spain according to EMEP(see Fig. 33 and 34) (they strated to decrease only recently)
- The decease reported by EMEP is substantially larger than the values of the TEAM model (comp. Fig. 27 and 28), showing that the uncertainties of such emission models are large.

Fig. 35: Measurement sites of the Swiss air pollutant monitoring network NABEL (Suter, 2011)

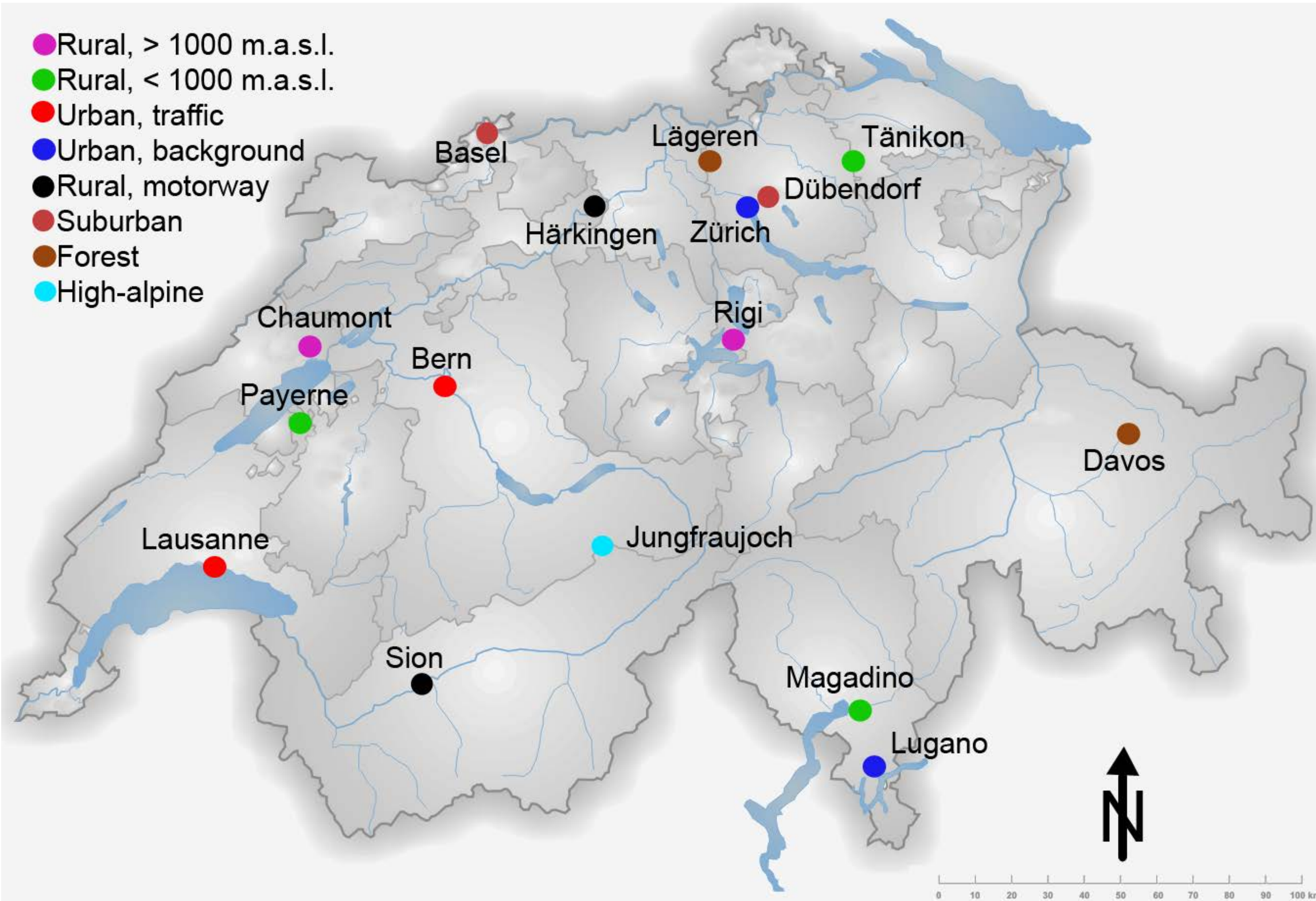
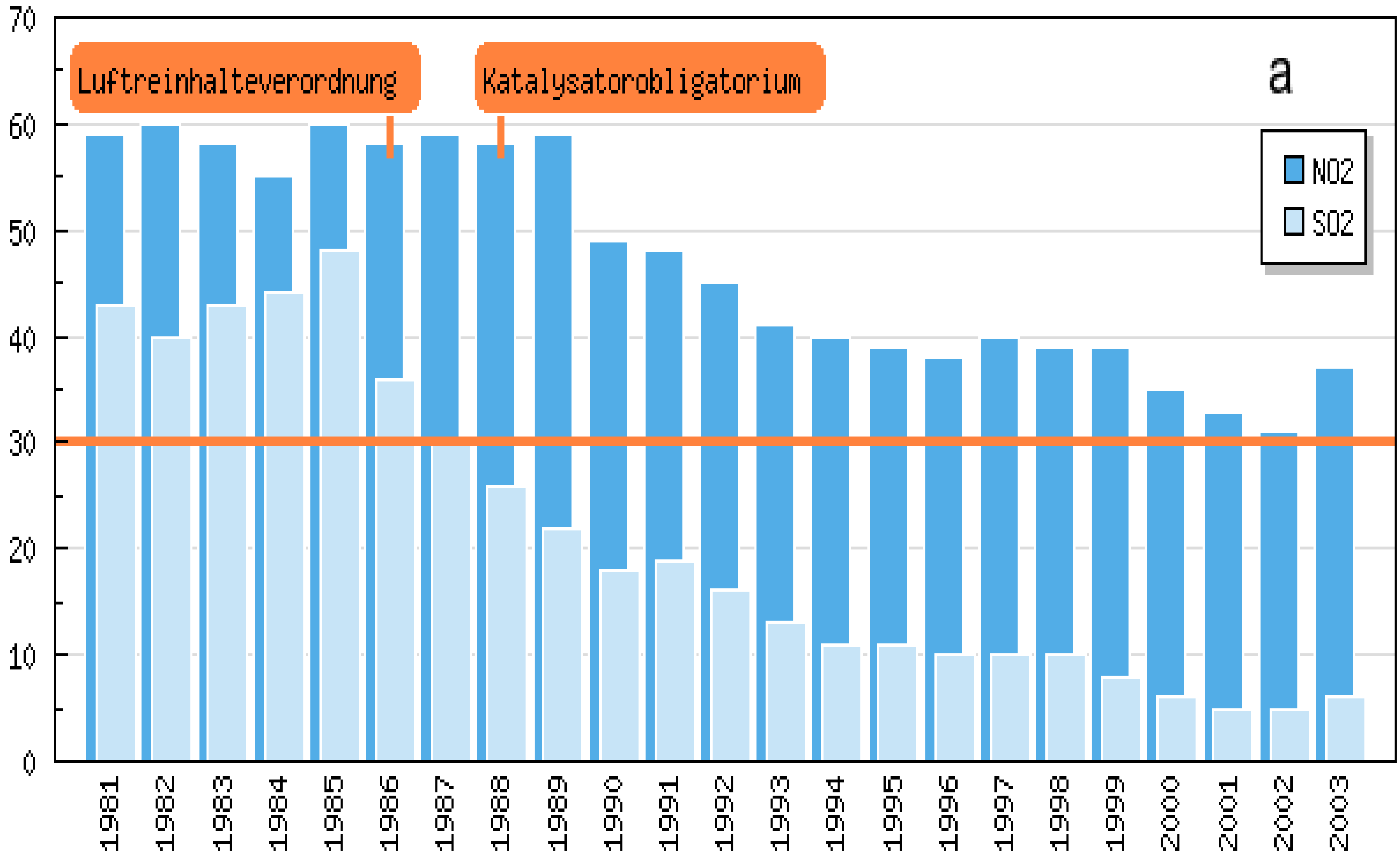


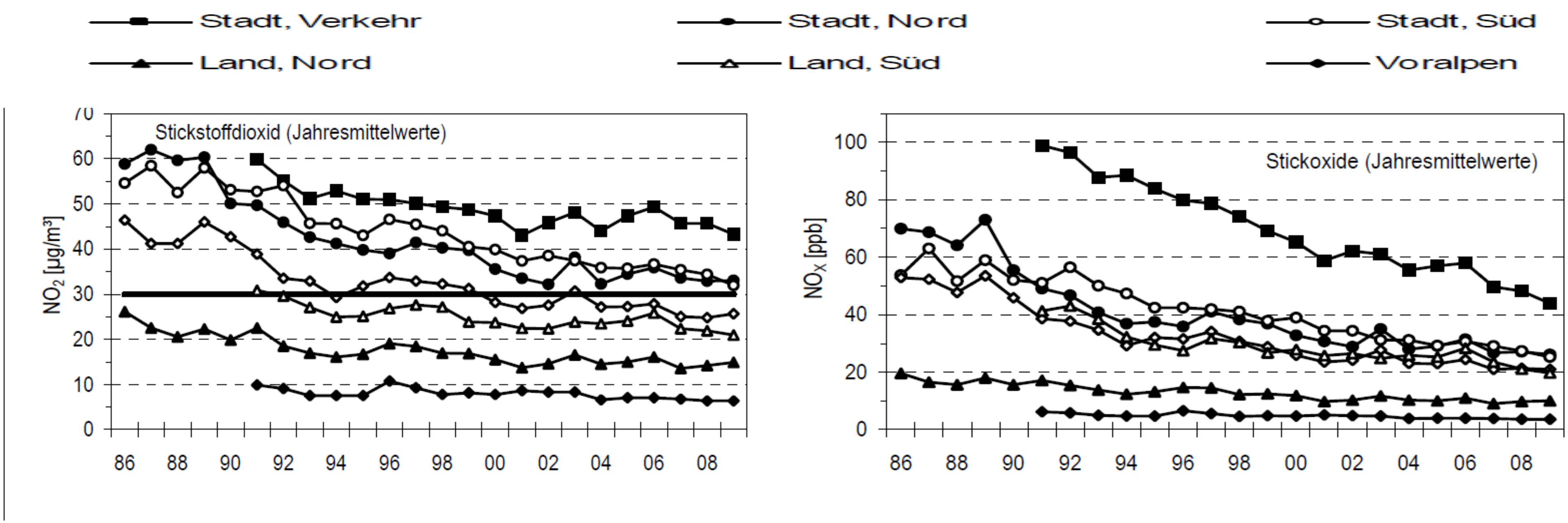
Fig. 36: Concentrations at Kasernenhof Zürich (NABEL: Nationales Beobachtungsnetz für Luftfremdstoffe, operated by EMPA)



Notes to Fig. 35 - 36: Swiss legislation and NABEL

- According to Swiss national legislation the Cantons need to monitor air pollutant concentrations of the compounds listed in Fig. 21. If the limiting values are exceeded in ambient air, the Cantons need to construct a plan, how to reduce air pollutants („Massnahmenplan“), which has to be approved by the Central Government in Bern.
- BAFU (Bundesamt für Umweltschutz, Environmental Protection Agency of Switzerland) funds (partailly) an additional air pollution monitoring network, which is operated by EMPA (NABEL: Nationales Beobachtungsnetz für Luftfremdstoffe) (see Fig. 35).
- The sites (see Fig. 35) are classified as: Urban traffic; urban background; suburban; rural; rural motorway (Härkingen), alpine foothills or Jura mountain; high Alpine site (Jungfraujoch)
- Longest time series of NABEL started at the early 1980s (Fig. 36), most (reliable and complete) series go back to the early 1990s. Fig. 36, 37 and 38 compare the measurements of primary air pollutants of NABEL with the (long-term) Swiss anthropogenic emission inventory.
- The emission inventory includes traffic, industry, households, agriculture and forestry. They were calculated for every 5 years.

Fig. 37: Changes in Swiss anthropogenic emissions and ambient air concentrations of NO_x (NABEL, 2010)



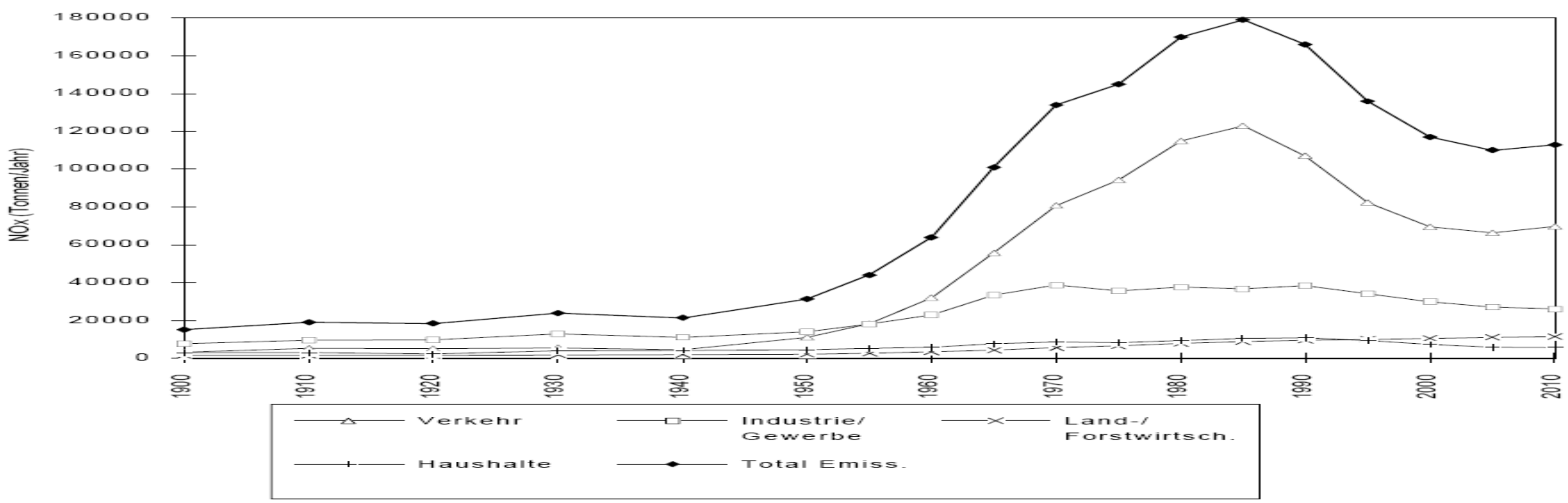
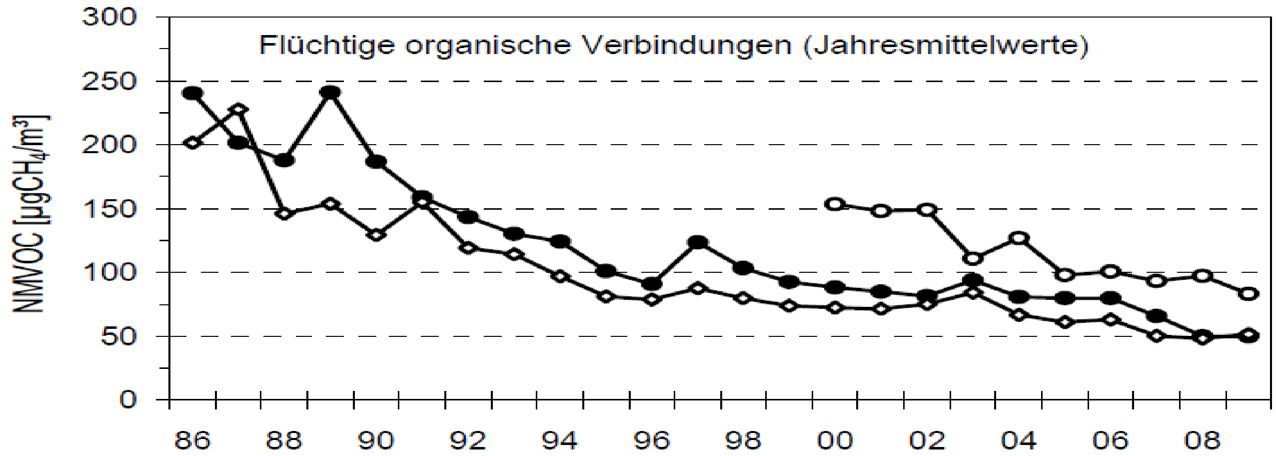


Fig. 4.2 Stickoxid-Emissionen 1900 - 2010

Notes to Fig. 37: NO_x changes in Switzerland

- (According to the governemental emission inventory) Swiss NO_x emissions increased by more than a factor of 4 between 1950 until 1985, mainly because of the very large increase in road traffic emissions.
- After 1985 road traffic emissions of NO_x strongly decreased (mainly because of the introduction of catalytic converters in gasoline driven vehicles).
- At the urban background site „Kasernenhof“ in Zürich (where also many additional air pollutant are measured) NO_2 concentrations strongly decreased in the years after 1989 (comp. Fig. 35). Note that catalytic converters were introduced in new gasoline driven cars in 1988.
- For 2000 to 2010 the NO_x emission inventory shows only small changes.
- Ambient air concentrations of highly polluted sites decreased during the 1990s, in agreement with the national emission inventory.
- Similar amount of decrease were measured at urban background, suburban and rural sites in Switzerland.
- NO_2 concentrations also decreased at polluted sites in Switzerland, but much less than NO_x.
- NO_2 annual limiting values are still exceeded not only at strongly polluted sites but also at urban background sites and suburban sites.

Fig. 38: Changes in Swiss anthropogenic emissions and ambient air concentrations of t-NMVOC (NABEL, 2010)



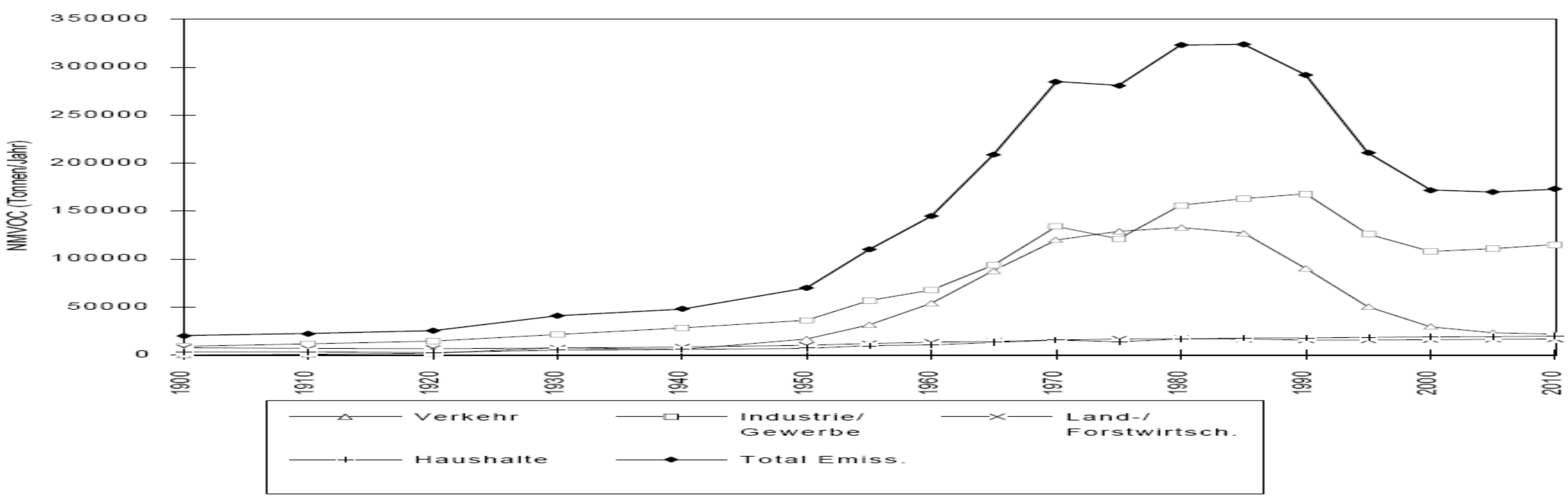
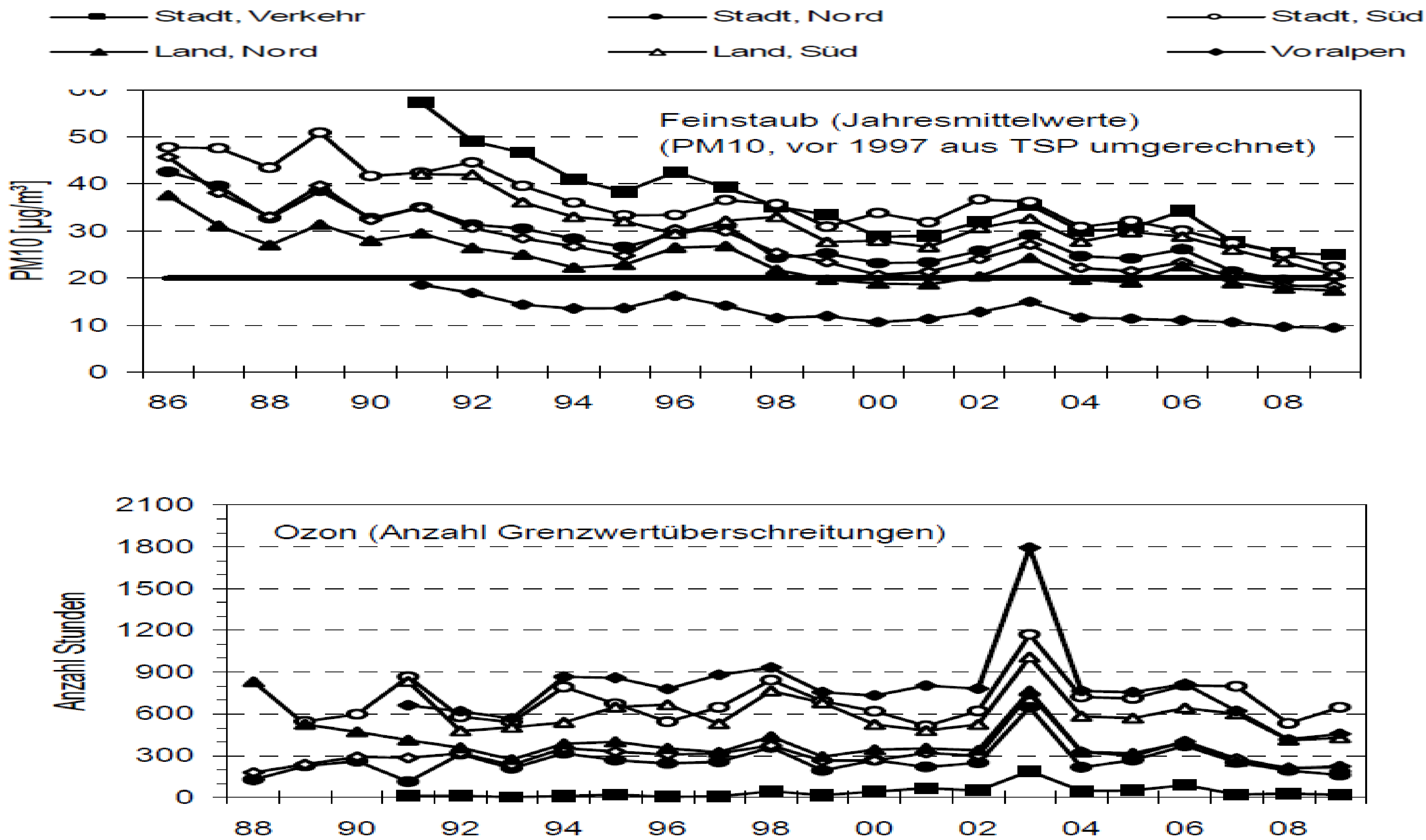


Fig. 4.3 NMVOC-Emissionen 1900 - 2010

Notes to Fig. 38: Changes in t-NMVOC in Switzerland

- (Total) t-NMVOC is only measured at 3 sites (one only since 2000) in NABEL using FID-detecors, which only provides an approximate information (because the individual VOCs have different repsonses to the detector).
- The temporal evolution of the anthropogenic t-NMVOC Swiss emission inventory is similar to the NO_x emissions.
- The most important t-NMVOC emission sources are (according to the inventory) industry and road traffic.
- According to the emission inventory t-NMVOC emissions strongly decreased since 1985 (manily because of changes in road traffic emission).
- Catalytic converters very efficiently decrease road traffic t-NMVOC emissions.
- The available measurements of t-NMVOC support the large decrease (by approximately a factor of 2) in NMVOC-emissions predicted by the emission inventory.

Fig. 39: temporal evolution of particulate matter and ozone (hours exeeding threshold) from NABEL, 2010



Notes to Fig. 39

- According to Swiss legislation PM10 (particlulate matter with aerodynamic diameter of maximally 10 μm) is measured since 1998 (see Fig. 21). The data prior to 1998 were calculated from TSP (Total Suspended Matter) measurements by conversion factor(s) deduced from simultanues PM10 and TSP measurements.
- PM10 measurements decreased during the early 1990s but the annual mean values at polluted sites are still much higher than the limit values (comp. Fig. 39).
- For ozone the Swiss legislation specifies a threshold value which is not allowed to be exceeded more than once (hourly value) per year (see Fig. 21). The number of hours of exceedances is larger South to the Alps than in the Swiss plateau and did not change much over time (Fig. 39).
- The lack of a decrease in ozone values above 120 $\mu g/m^3$ is surprising keeping in mind the large decrease in ozone precusor concentrations (Fig. 37 and 38).
- Ozone maxima only marginaly decreased in the Swiss plateau and somewhat more in the Ticino area, which is more influenced by the polluted Milan area (see NABEL, 2010).
- Fig. 39 (bottom, hours exceeding the threshold of Swiss National legislation for ozone) illustrates the importance of meteorology on air pollutant concentrations (remember that summer meteorology of 2003 was characterized by the “heat wave”).
- Statistical models can be applied to attribute (remove) part of the meteorological variability using appropriate meteorological proxies (covariates).
- This procedure is called “meteorological adjustment”, yielding concentrations for meteorologically averaged conditions - for ambient concentrations of a particular air pollutant at a particular site.
- NABEL measurements treated by meteorological adjustment are used for longterm trend analysis in Fig. 40 - 43.

Fig. 40: Mean annual relative linear trends of meteorologically adjusted **NOx** concentrations at 13 NABEL stations compared to the linear trend fit of the emission inventory of EMEP (1990-2009). Red: highly polluted urban sites; blue: close to motorway; black: sub-urban or urban background; green: rural sites; Lugano dashed: south of the Alps (the results are grouped according mean concentrations, largest left)

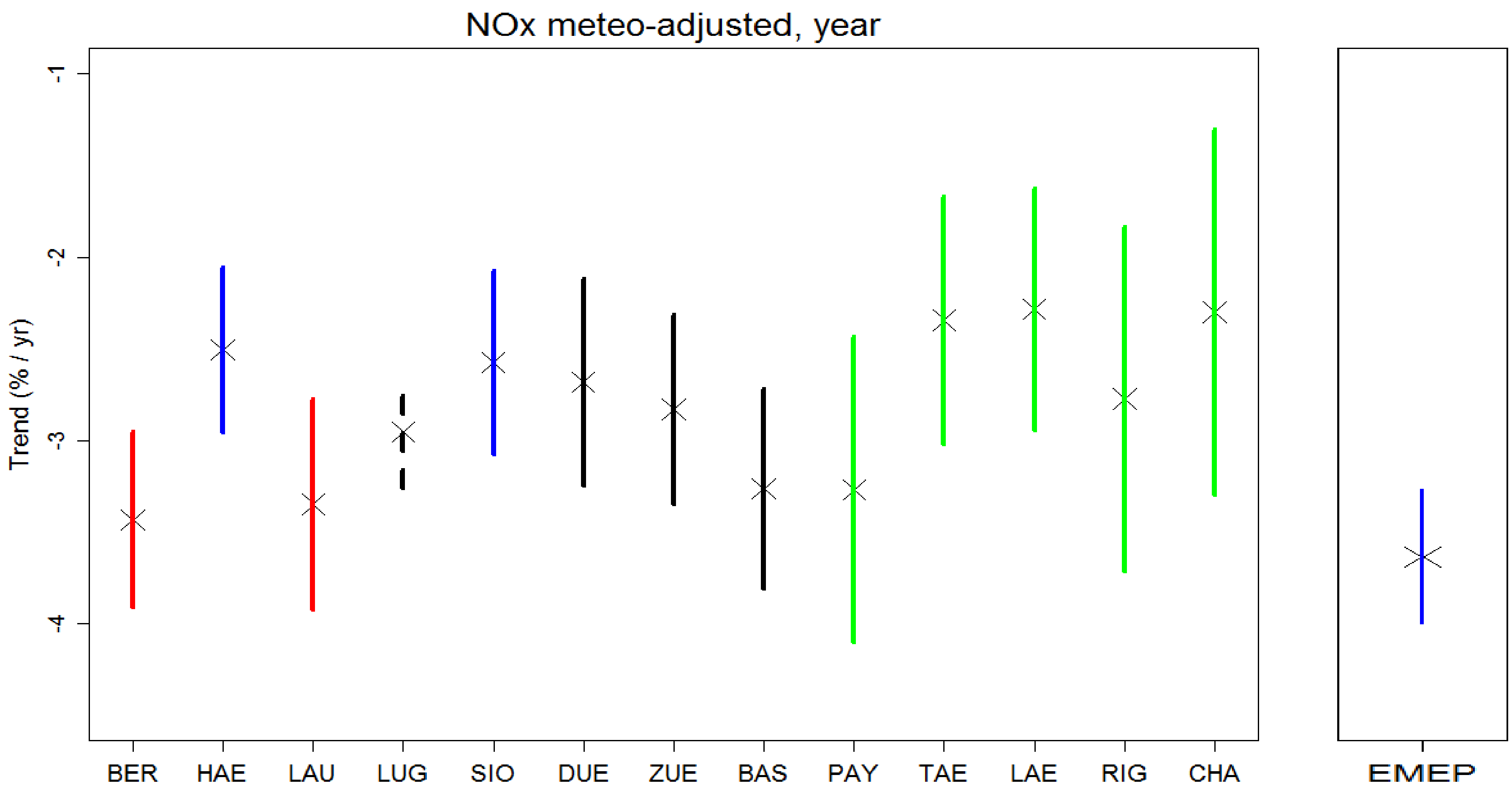
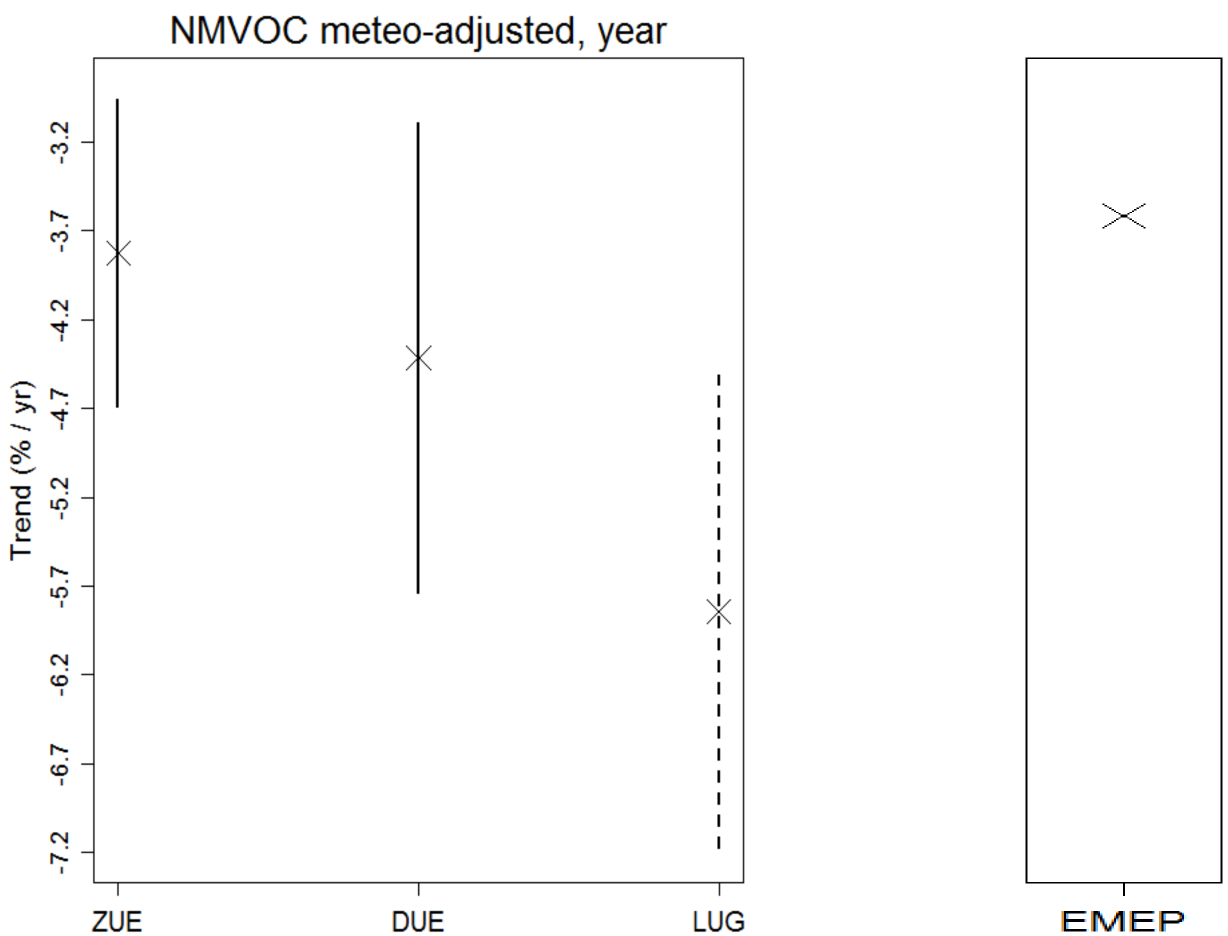


Fig. 41: Mean annual relative linear trends of meteorologically adjusted NMVOC concentrations at three NABEL stations compared to the decrease calculated from EMEP NMVOC emission inventory. DUE 1990-2009, ZUE 1990-2008, LUG 2000-2009, EMEP 1990-2010.



Notes to Fig 40 - 43: Meteorological Adjustment of NABEL ambient air measurements

- Suter (2010) applied Generalized Additive Models (GAM) for meteorological adjustment of long-term NABEL measurements
- Such GAM models have the following mathematical form

$$C_i = \sum_{j=1}^{8} s_j(x_i) + a + \epsilon_i$$

Whereas: C_i: observation *i* of concentration of air pollutant C; $s_j(x_i)$: smooth function of variable *j* for observation *i*; (*a*: constant, not used in this study); $\mathcal{E}_i$: residual of observation *i*

- GAM models were applied for every station and all studied pollutants
- target values (summer data) were: daily means for NOx and t-NMVOC, daily maxima for O_3 and Ox (NO_2 + O_3) and the ten highest daily O_3 and Ox concentrations per summer season (MTDM: **m**ean of **t**en **d**aily **m**axima)
- In order to describe the meteorological influence on air pollutant concentrations 61 meteorological variables (e.g. afternoon temperature, local temperature, synoptic daily classification) were tested as model predictors
- The first step was to select the meteorological covariates which have largest effect of pollutant concentrations at the sites; for this purpose forward selection was used
- Trend estimate: several approaches were tested giving very similar results
- In the final models 4-5 meteorological predictors were used (explaining most of the variability)
- For summer daily O_3 maxima most often selected variables were: afternoon temperature; global radiation; water mixing ratio or relative humidity; wind direction and or wind speed, day of the week; days after frontal passage

Notes to Fig. 41 and 42: Trends of meteorologically adjusted of ozone precursor concentrations

Large decreases in ozone precursor concentrations were found:

- NOx decrease (Fig. 40) was equivalent to a total decrease of about 55% for NOx (1990-2009)
- NOx decreases are were found to be largest at polluted sites (possibly because at less polluted sites NOx background concentrations are more important)
- t-NMVOC (Fig. 41) was large (at the few sites): around 60%; at Lugano the decrease was even larger than in the EMEP emission inventory, possibly because of decrease in t-NMVOC concentrations importet from the Milan area
- The decreases deduced from meteorologically adjusted NABEL ambient air measurements were similar to the EMEP emission inventory and therefore support the Swiss national emission inventory (reported to EMEP);
- The large decreases in primary pollutant concentrations confirm the effectiveness of air pollutant control measures started in the second half of the 1980s (e.g. introduction of catalytic converters in new gasoline driven cars)

Fig. 42: Mean relative trends of summer daily maximum of ozone concentrations (top) and of Ox concentrations (bottom) at NABEL stations (1990-2009). X: trends estimated from measured data, □: trends calculated from meteorologically adjusted data (grouped according NOx mean concentrations, highest concentrations left) (Suter, 2011)

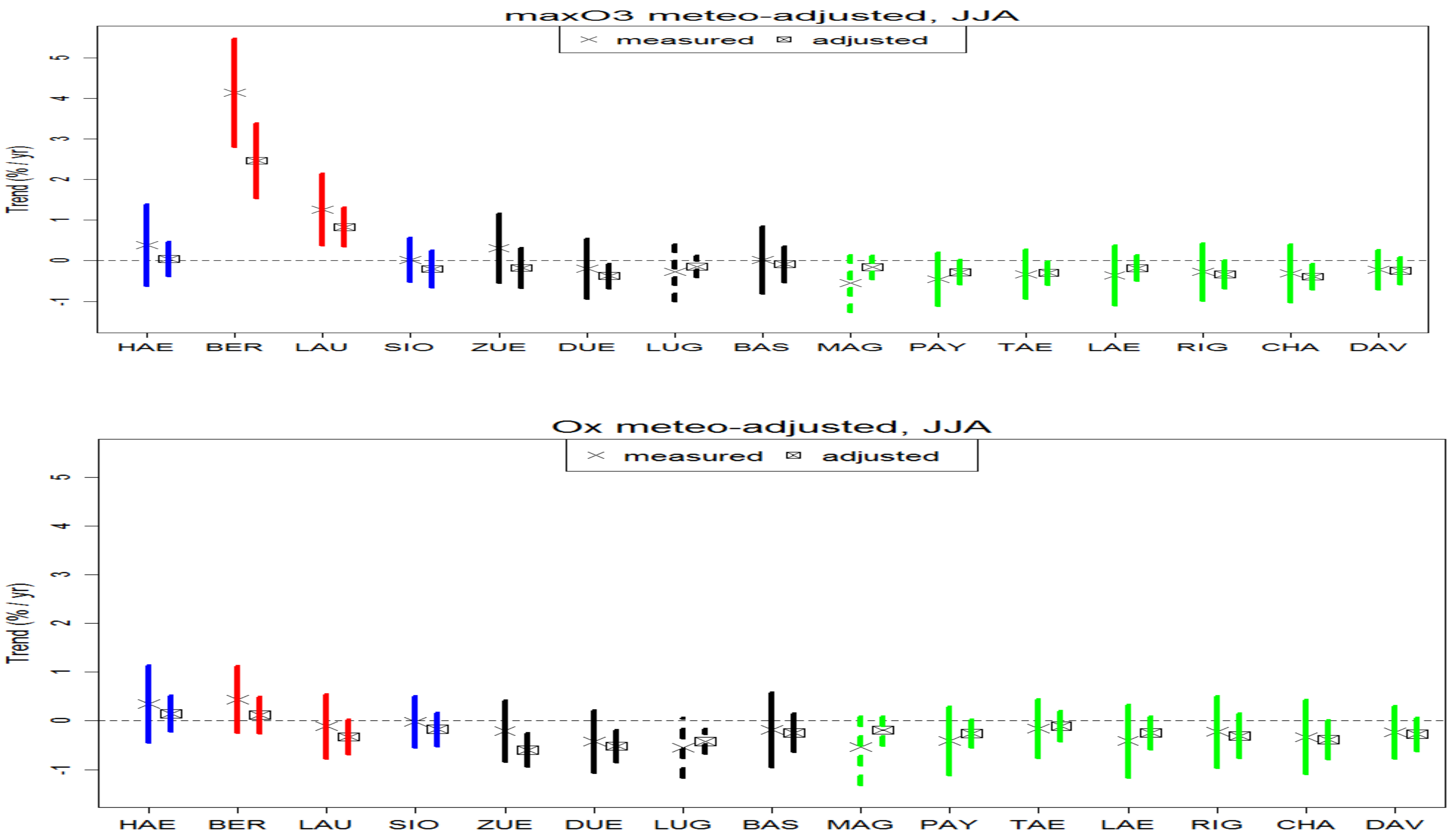
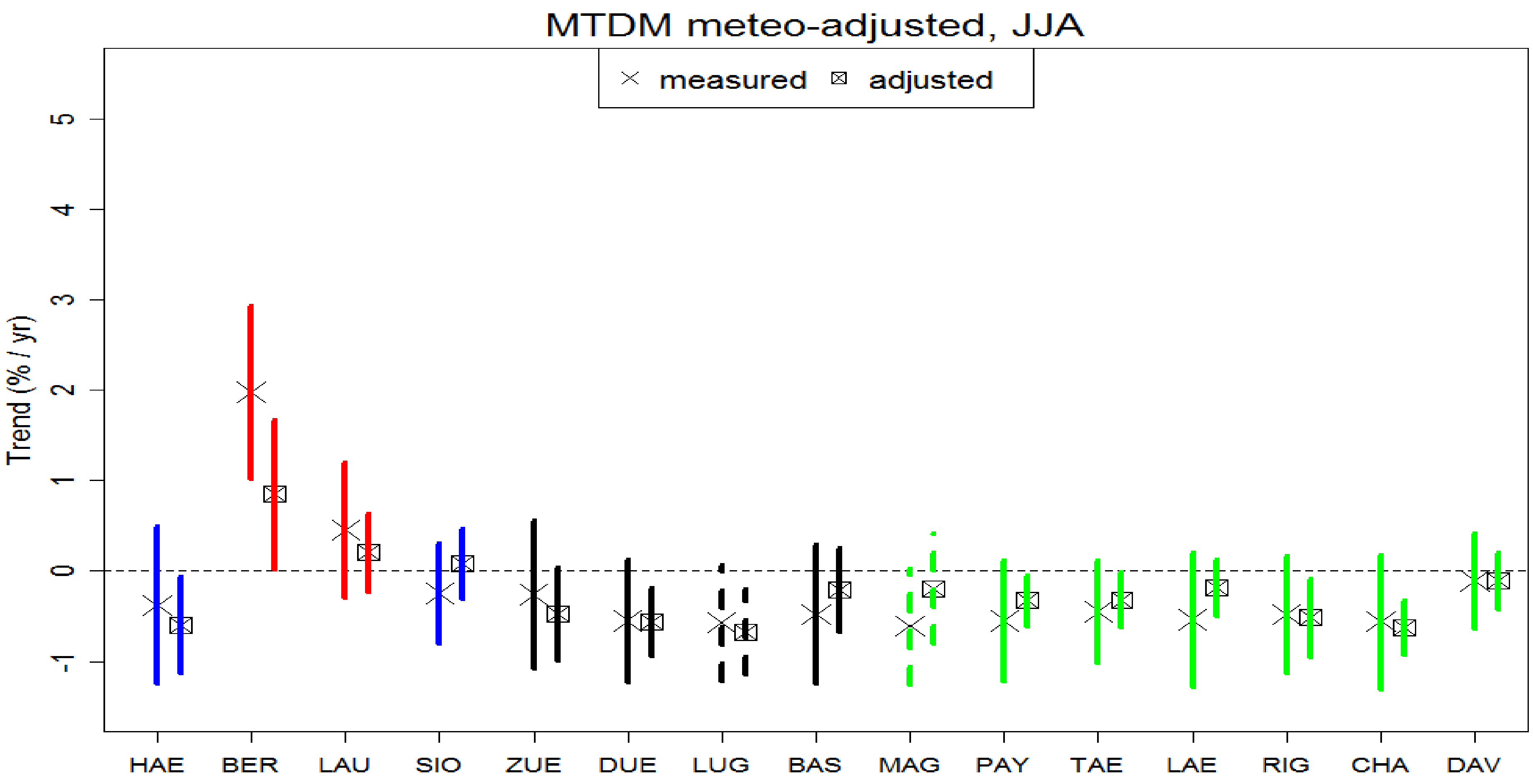


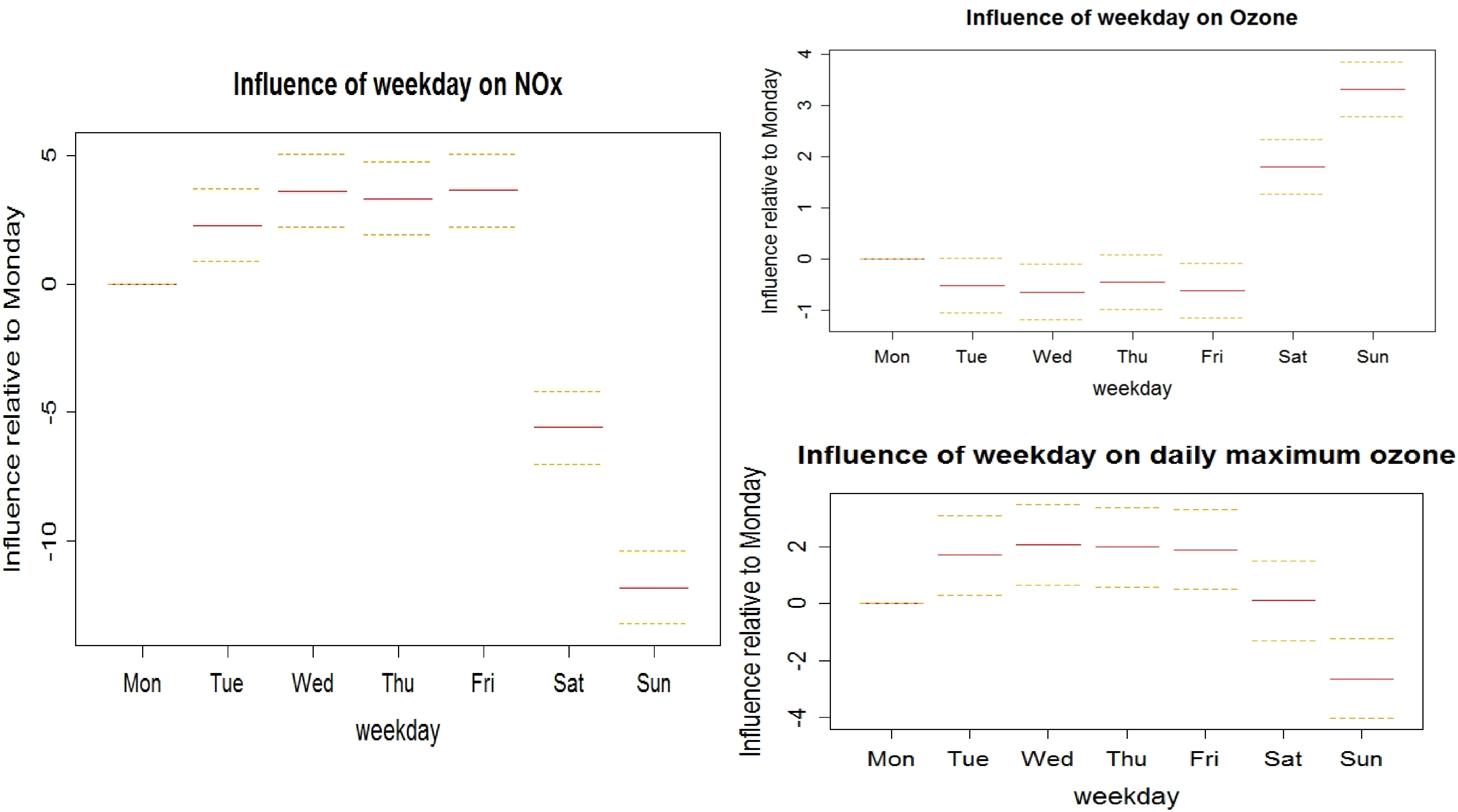
Fig. 43: Mean relative trends of the mean of the ten highest daily maximum ozone concentrations (MTDM) at NABEL stations (1990-2009). X: trends estimated from measured data, □: trends calculated from meteorologically adjusted data (grouped according NOx mean concentrations, highest concentrations left) (Suter, 2011)



Notes to Fig. 42 and 43: Ozone trends from NABEL measurements

- Ozone: Significant increases were found at urban polluted sites (explicable by titration, i.e. reaction between NO and O_3)
- Ox values: No upward trends were found at polluted sites, confirming importance of titration (Fig. 42)
- Rural PBL sites: Ozone and Ox trends: less positive/more negative (similar results were obtained by an other statistical model approach (Ordonez et al., 2005))
- Meteorological adjustment mainly leads to smaller uncertainties in trend estimates whereas trend magnitude was similar (Fig. 42 and 43)
- The trends in meteorologically adjusted rural summer maxO3 (~-4%) and MTDM (~-7%) were found to be statistically significant at most non urban sites but they are unexpectedly small keeping in mind the large decrease in ozone precursor concentrations
- The EMEP Unified Chemical Transport Model model was used to describe the evolution of ground level ozone concentrations from 1995-2005. EMEP model uses a 50*50km grid; in the numerical simulations meteorology as well as the decrease in emissions of ozone precursors (all over Europe) was described. The EMEP model predicts decreases of -25% of MTDM for Swiss rural sites (1995-2005), much larger than found in the meteorologically adjusted measurements
- The difference might be caused by downward mixing of background air (see 4.10, Fig. 45)

Fig. 44: Influence of weekday on air pollutant concentrations (1990-2009) determined after fitting GAM models: left: mean daily NO_x concentrations in Zürich; right: top: summer mean daily ozone concentrations in Zürich.; bottom: summer daily ozone maximum concentrations in Lägeren (Suter, 2011)

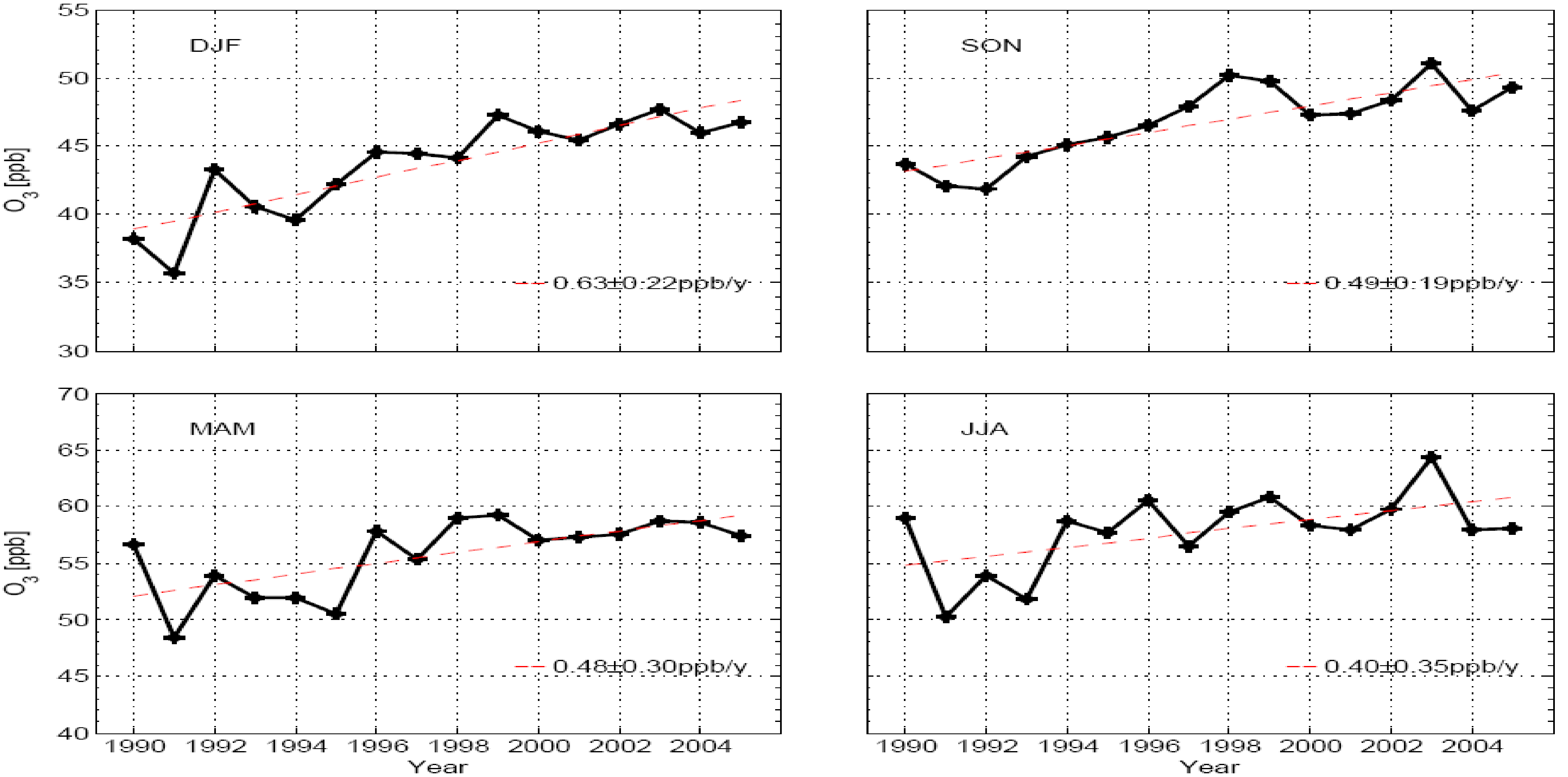


Notes to Fig. 44: “Weekend effect”

- During weekend good transport is forbidden in Switzerland and therefore NO_x emission strength is considerably larger during the working days; this is shown by the NO_x measurements at Zürich treated by the GAM model (see Fig. 44). (The small increase from Monday to Wednesday might be caused by accumulation of NO_x)
- At the polluted site Zürich summer mean daily ozone concentrations are larger on weekend explicable by titration (when NO emissions are larger (on weekdays) less O_3 is destroyed and therefore O_3 concentrations are larger during weekend
- The weekend effect on O_3 has a different sign at rural sites than at urban (polluted sites) (comp. the two Fig. on the right side of Fig. 44): At the rural site Lägeren O_3 (summer daily maxima) are larger during the working days than during weekend: when an airmass is moved from a polluted site (where titration is dominant) O_3 is formed by photochemistry during the transport of the air parcel to a rural site leading to opposite sign in O_3 response to NOx emission

4.10. Ozone changes in Northern mid-latitudes

Fig. 45: Ozone changes at Jungfraujoch (Cui et al., 2011)



Notes to Fig. 45

- Ozone at the high Alpine site Jungfraujoch was increasing since the early 1990s (as first described by Brönnimann et al., 2002) until around 2000
- Ozone increase was similar at three high alpine sites (Jungfraujoch, Sonnblick (Austria), Zugspitze(Germany) between 1992 and 2002, the largest increase was found in winter (Ordonez et al., 2007).
- Also at the Mace Head, a coastal site in West Ireland, ozone was increasing, most strongly in winter. For the period 1987-2003 ozone increase amounted to (in $ppby^{-1}$): winter: 0.63; summer: 0.39 (Simmonds et al., 2004)
- This observation is surprising considering the substantial decrease in anthropogenic ozone precursor emissions in Europe in the same period.
- The increase in ozone background might have compensated (during the 1990s) the expected decrease in ozone maxima in the Swiss planetary boundary layer (comp. Fig. 42)

Fig. 46: Potential causes for background ozone increase over Europe: NO_2 tropospheric column increase over East and Central China (from satellite observation) (Richter et al., 2005)

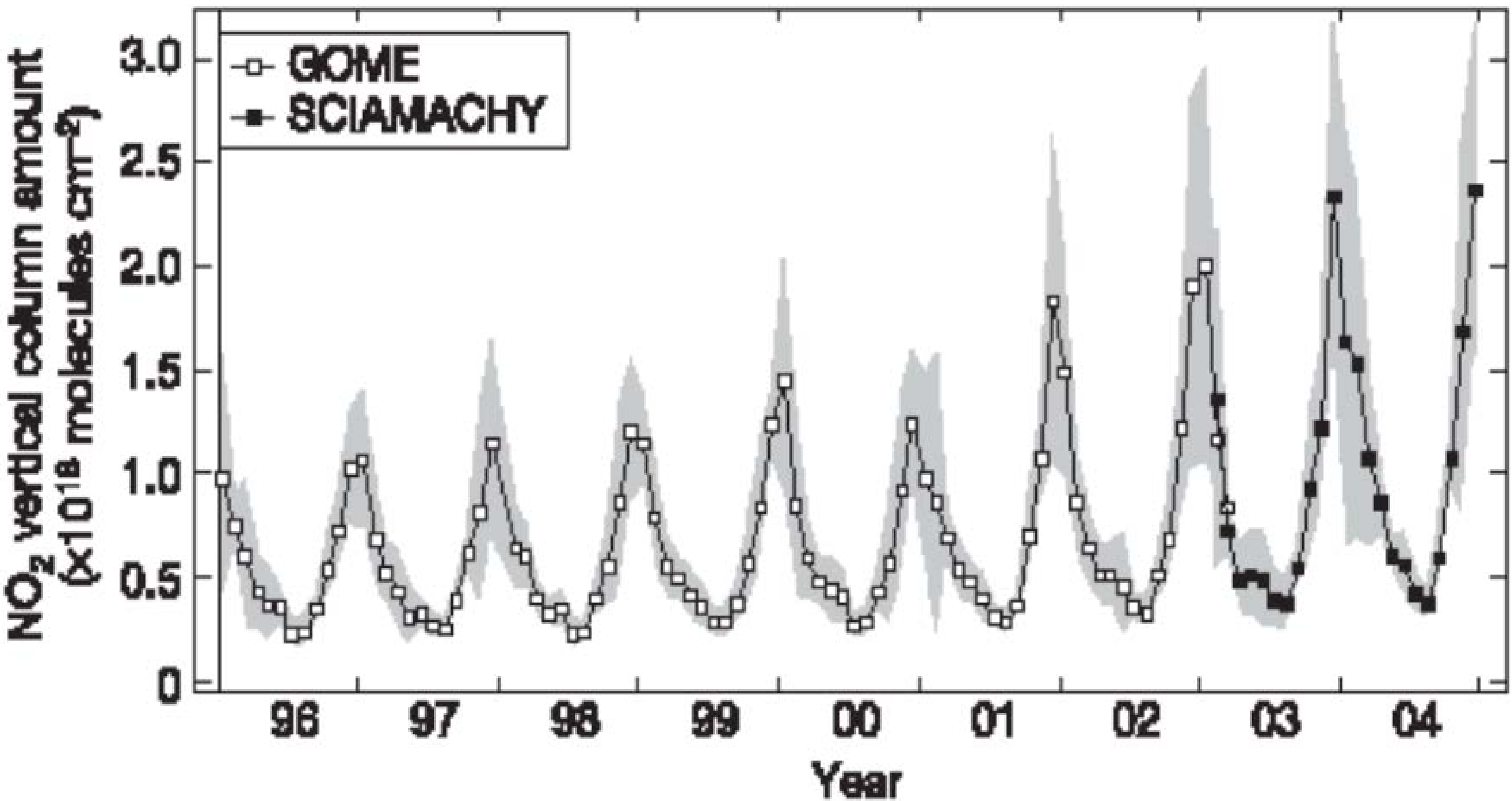
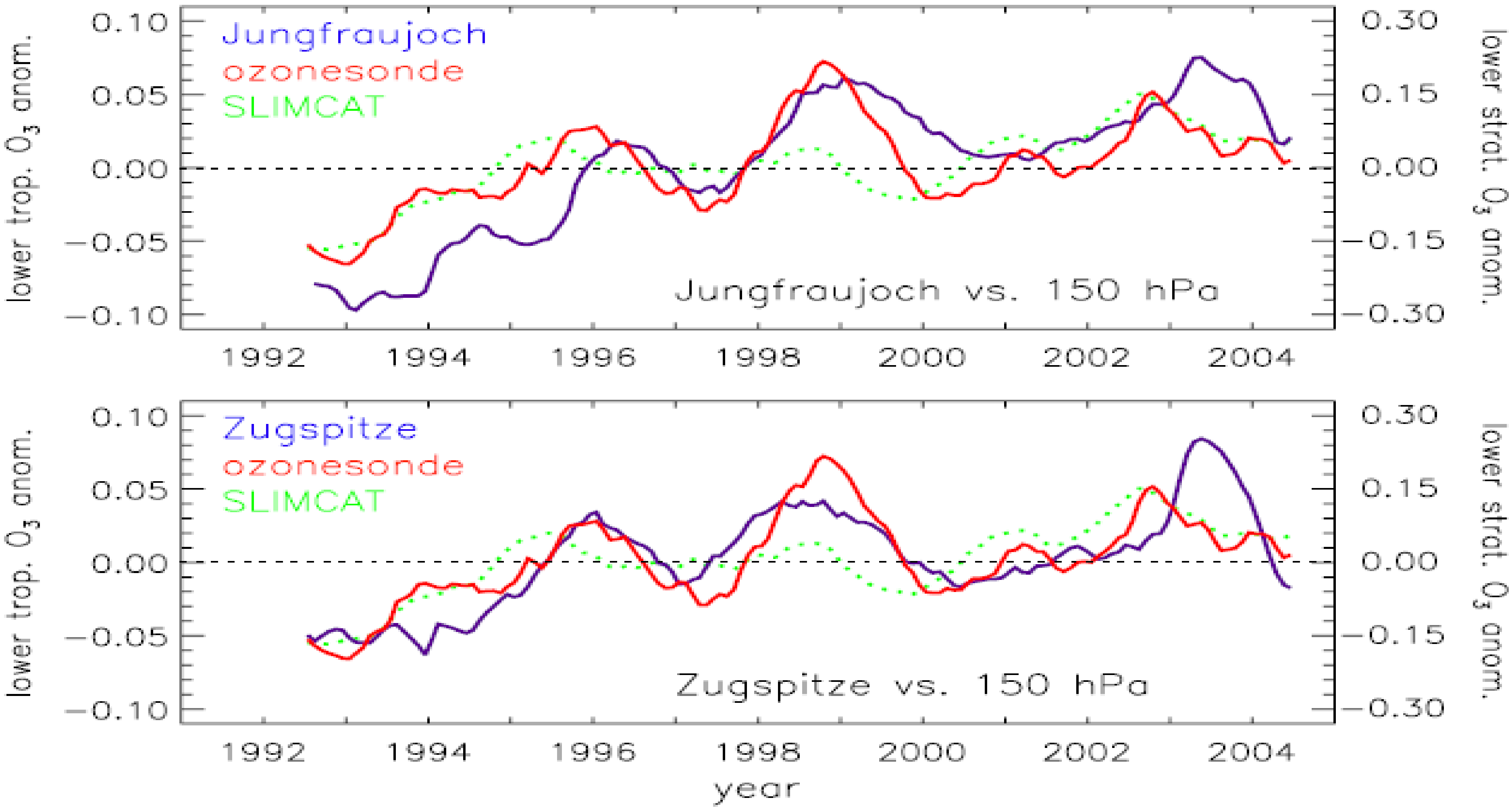


Fig. 47: Ozone change in the stratosphere (Ordonez et al., 2007)



Notes to Fig. 46 and 47

Two potential causes for the unexpected increase in background ozone which took place in the 1990s (as found at Jungfraujoch, see Fig. 45) were discussed:

- Large increase in **ozone precursor emissions of East Asia** (China) (Fig. 46)
- The increase of Asian ozone precursor emissions most probably caused a long-term increase in background ozone concentrations at the West coast of North America (see Cooper et al., 2010) but the Jungfraujoch increase is hardly attributable to East Asian emission increase, which did not stabilize around 2000.
- **Change in flux of ozone mixed down from the stratosphere** (Fig. 47). Part of tropospheric ozone originates from the stratosphere. Low ozone values occurred in the lower stratosphere in the early 1990s which was attributed to the effect of the large volcanic eruption Pinatubo. Fig. 47 compares the monthly mean anomalies of the ozone concentrations in the lower stratosphere (deduced from the ozonesondes of Payerne, Swiss Plateau (upper panel) and Hohenpeissenberg, Southern Germany (lower panel)) with those of the ozone measurements of the high alpine sites Jungfraujoch (upper panel) and Zugspitze (lower panel). The similarities of the time evolution of the anomalies of the stratospheric and tropospheric ozone provides qualitative evidence that the ozone background increase observed during the 1990s (measurements of Jungfraujoch) can be attributed to the ozone evolution in the lower most stratosphere (i.e. transport from the stratosphere).

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