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## Data analysis and testing of parameterisations

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### Summary

Estimates of diurnal and seasonal variation of ozone flux parameters to a temperate coniferous forest over 3 years in Southern Norway are presented. Based on available measurements from a weather and a sonic station the turbulent transfer coefficient, the ozone flux and the deposition velocity are calculated by the gradient method using alternative profile parameterisations. Corrections for the surface roughness layer are implemented by multiplying the fluxes with an enhancement factor of 1.42 estimated from characteristically length sizes of the site studied.

Even if annual and diurnal profiles of directly measured meteorological and sonic parameters show well known patterns, the corresponding resulting curves for the ozone flux and the deposition velocity are highly variable with unexpected elevated values during night, especially in fall. The transfer coefficient demonstrates a characteristically diurnal pattern with a midday peak for all seasons and lowest specification in winter. Performances with alternative profile parameterisations and a comparison with a sonic transfer coefficient correspond very well, particularly in the timing of the profiles. The noisy annual and diurnal cycles of the ozone flux and deposition velocity are thought to be a consequence from the poor data base resulting out of malfunction of the measurement stations and data processing.

Improved results are expected if longer time series are considered in the calculations of the daily and monthly averages. Additionally the elevation of the upper measurement height is thought to ameliorate the measurements and data base when calculating the deposition flux with the gradient method.

## **Summary in Norwegian**

Estimater av daglige og årlige variasjoner i ozonavsetning til barskog i Hurdal blir presentert for perioden 2004-2006. Basert på tilgjengelige målinger fra værstasjon og sonisk anemometer har den turbulente transfer coefficient, overføringskoeffisient, deposisjonshastighet og ozonfluks blitt beregnet med forskjellige parameteriseringer fra litteraturen. Det har blitt korrigert for effekter av nærhet til en ru overflate ved at det anvendes er korreksjonsfaktor på 1.42 estimert ut fra karakteristiske lengdeskalaer.

Selv om årlige og daglige variasjoner av meteorologiske parametere viser forventet utvikling, samsvarer ikke ozonfluksen med tilsvarende observasjoner fra litteraturen. Særlig er det uventet høye verdier om natten og om høsten. Den turbulente overføringskoeffisienten har en forventet utvikling med maksimum midt på dagen og lavest verdi om vinteren. Forskjellige parameteriseringer og sammenligning mellom soniske og tradisjonelle meteorologiske målinger sammenfaller bra, særlig i timingen av profilene. De uventede resultatene for ozonfluksen kan delvis skyldes manglende datadekning i perioder.

Forbedrede resultater kan forventes hvis lengre tidsserier blir brukt i beregningen av middelverdiene. I tillegg er masten trolig for lav til å få fullverdige målinger med gradientmetoden.

## Ozone deposition in Hurdal 2004–2006: Data analysis and testing of parameterisations

#### **1** Introduction

Quantification of ozone deposition to ecosystems is important for many reasons, including developing and validation of models for regional deposition and estimating the vegetation exposure to ozone (Keronen et al., 2003; Hole et al., 2004; Mikkelsen et al., 2004; Tuovinen et al., 2004; Karlsson et al., 2006). To establish a new generation of such complex modelling tools with the ability to provide detailed deposition maps, knowledge about characteristic seasonal and diurnal cycles of ozone fluxes and deposition velocities will be crucial (Emberson et al., 2000; Simpson et al., 2002). Wesely and Hicks (2000) have described dry deposition processes in general and the current status of knowledge in the field in an extensive review. Several authors have reported measured and modelled ozone deposition to coniferous forests in Europe (e.g. Pilegaard et al., 1995; Zeller, 2002; Lamaud et al., 2002; Hole et al., 2004) and comparisons between different flux calculations have been presented (Mikkelsen et al., 2000; Keronen et al., 2003; Mikkelsen et al., 2004).

In this report we calculate annual and seasonal diurnal cycles of the ozone deposition flux above a coniferous forest in Southern Norway under continental Scandinavian conditions. Patterns of relevant meteorological parameters as well as the ozone gradient and concentration measurements are presented and the roughness length for the spruce forest is calculated in order to characterize the site. Turbulent transfer coefficients, deposition velocities and ozone flux at 20 m above ground are assessed by the gradient method. Contrary to earlier calculations performed for the same site by Hole et al. (2004) for the period between 2000 and 2003, additional data from a recently installed sonic anemometer enabled a broader range of available input variables. By using different parameterisations reported in literature and correcting the results with an enhancement factor alternative calculation methods are compared and the quality of the measurement setting is evaluated.

Our main objective in this study is to determine whether installation of the sonic anemometer in Hurdal has improved the quality of the flux measurement or whether further changes in the measurement set-up is required.

#### 2 Site description

Since 1 July 2000 monitoring of ozone deposition has been undertaken by the Norwegian Institute for Air Research (NILU) in a Norway spruce forest (*Picea abies*) in Hurdal, South-East Norway (60°22'N, 11°4'E). The data presented here are from the period 1 January 2004 to 31 December 2006. The area is relatively homogeneous in tree height and topography and was clear-cut about 40 years ago. Average tree height is now around 13 m. The entire region is generally covered with spruce forest mixed with farm land and lakes and the fetch is forested for

several kilometres in all directions. The climate is typical for the Scandinavian interior with winter January normal temperature of -7.2 °C and July normal temperature of +15.2 °C.

#### 3 Instrumentation and data processing

Wind and air temperature profiles are measured using a 25 m tower. Temperature difference between 15 m and 25 m, wind speed and direction at 25 m, and relative humidity at 25 m is averaged and sampled every hour by an Aanderaa weather station. The temperature difference has an accuracy of 0.1 K (or 0.01 Km<sup>-1</sup> here), while average wind speed measurements have an accuracy of 0.2 ms<sup>-1</sup>. Wind speeds below 0.5 ms<sup>-1</sup> are considered calm to avoid problems with the sign of the wind speed gradient. Ozone concentrations at 15 m and 25 m above ground are measured by an UV absorption probe (API 400 O<sub>3</sub> analyser), switching from intakes at the two heights in 5-min cycles. All weather station and ozone data are stored as 1 hour averages.

Additional data is provided by a sonic anemometer ("Sx" Style probe) measuring wind velocity components in three axis and the sonic temperature of the winds (Applied Technologies Inc., 2007). While the wind speed in all directions has an accuracy of 0.03 ms<sup>-1</sup>, the sonic temperature has an accuracy of 0.1 °C. Sensible heat flux and friction velocity are derived from these data and stored as half hourly averages. Thus an interpolation of the sonic measurements has been made, in order to combine the data from the two different instrumentations in the flux calculations.

The raw data have been corrected from noise and blind values using reasonable limits. In the records of the ozone concentrations, the wind speed and the friction velocity all values below zero were removed. Further limitation of the friction velocity was performed later in the calculations as it is described in the next chapter. Temperatures above 40 °C were treated as erroneous data as well as values below -40 °C. Taking into account the geographical position of the measurement site, sensible heat fluxes below -70 Wm<sup>-2</sup> and above 1000 Wm<sup>-2</sup> were considered as noise. Averaged over all years and all parameters 5% of the data were removed through this procedure, whereas the percentage of the loss ranges from 0 for the temperature in all years to 15 for the friction velocity in the year 2006. This value already includes the additional limitation of the parameter throughout the calculation, thus the higher loss factor can be explained. Figure 1 shows the resulting workdata that served as input into the flux calculations.

In the calculation of mean values each year and each hour in the day is given the same weight, even though the percentage of missing data for every season varies between 0 and 80 depending on the parameter. Affected from high loss rates are the parameters which are calculated from diverse datasets with lacking values during different months, namely the transfer coefficient, the ozone flux and the deposition velocity. Especially in winter and fall the missing sonic data lead to this small percentage of available values.

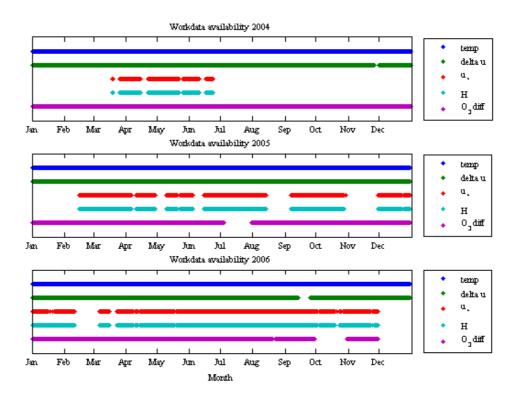


Figure 1: Availability of the workdata in the three measurement years. temp = Temperature, delta u = windspeed gradient between the measurement heights,  $u_* =$  friction velocity, H = sensible heat flux,  $O_3$ diff = ozone gradient between the measurement heights.

#### 4 Methods

#### 4.1 Flux calculations

The vertical ozone flux, F, and the deposition velocity,  $V_d$ , have been estimated by the gradient method following the recommendations of Fowler and Duyzer (1989) and Hummelshøj (1994) using two different parameterisations for the stability functions proposed in literature (van Ulden and Holtslag, 1985; Mikkelsen et al., 2004). Additional corrections for the roughness surface layer are implemented and described in chapter 4.3. The deposition velocity was calculated at 20 m above ground (i.e. in the middle of the two measurement heights  $z_1$  and  $z_2$ ) from the following equations:

$$F = -\frac{ku_*[c(z_2) - c(z_1)]}{\ln\frac{z_2 - d}{z_1 - d} + \psi_h((z_1 - d)/L) - \psi_h((z_2 - d)/L)} = -f[c(z_2) - c(z_1)]$$
(1)

$$V_d(z) = -\frac{F}{c(z)} \tag{2}$$

where k is the von Kàrmàns constant (=0.4),  $u_*$  is the friction velocity, c(z) is the ozone concentration at the level z, d is the displacement height, L is the Monin-Obukhov length and  $\psi_h$  is the integral form of the Monin-Obukhov stability

function for heat. The parameters determined from the vertical wind and temperature profiles can be summarized in the turbulent transfer coefficient f. The displacement height for the dense forest is set to 8 m using the common approximation  $d \approx 2/3 h$ , where h denotes the canopy height (Garratt, 1994). Contrary to some previous studies (e.g. Mikkelsen et al., 2000; Hole et al., 2004)  $u_*$  was directly measured by the sonic anemometer and not obtained through profile assumptions. The Monin-Obukhov length was determined from both sonic and weather station data using the equation

$$L = \frac{u_*^2}{k\frac{g}{T}\theta_*}$$
(3)

Here T is the absolute temperature and  $\theta_*$  is a turbulent temperature scale calculated by

$$\theta_* = -\frac{H}{\rho c_p u_*} \tag{4}$$

with *H* denoting the sensible heat flux,  $c_p$  the specific heat of air at a constant pressure (1006 Jkg<sup>-1</sup>K<sup>-1</sup>) and  $\rho$  the air density set to 1.12 kgm<sup>-3</sup>. Friction velocities below 0.05 ms<sup>-1</sup> are not considered in the calculations, for Monin-Obukhov similarity works only when the wind speeds are not calm (Garratt, 1994).

The first parameterisation of the stability functions for heat used in this report is presented by Mikkelsen et al. (2000), who calculated the ozone flux over a spruce forest situated in Denmark, a region comparable to the measurement site in Hurdal. Based on previous studies (Businger et al., 1971; Jensen et al., 1984; Högström, 1988) the following estimation is proposed:

$$\psi_h = -a\frac{z-d}{L} \qquad \qquad L > 0 \tag{5}$$

$$\psi_{h} = \left(1 - \beta \frac{z - d}{L}\right)^{1/4} - 1 \qquad L < 0 \tag{6}$$

where the values of  $\alpha$  and  $\beta$  are set to 8 and 12. In this formulation the integral form of the stability function for the stable case is calculated from the log-linear approximation and for the unstable case it is derived from the approximation  $\psi_h = \phi^{-1} - 1$ , where  $\phi$  is the dimensionless gradient function for wind shear. Van Ulden and Holtslag (1985) use the log-linear approximation also for the cases where L < 0, leading to the equations:

$$\psi_h = -\alpha \frac{z - d}{L} \qquad \qquad L > 0 \tag{7}$$

$$\psi_{h} = 2\ln\left(\frac{1 + \left(1 - \gamma \frac{z - d}{L}\right)^{1/2}}{2}\right) \qquad L < 0$$
(8)

This approach is the most common in literature dealing with flux-profile relationships and there has been a wide discussion about the parameters  $\alpha$  and  $\beta$  in the past. Several authors present a good overview of the suggested experimental values of the constants appearing in the stability functions (e.g. Dyer, 1974; Högström, 1988; Garrat, 1994). Van Ulden and Holtslag (1985) set the values of  $\alpha$  and  $\gamma$  to 5 respectively 16 following the recommendations of various previous authors (e.g. Dyer, 1974; Businger et al., 1971). Dyer (1974) considered these values as the most convincing at his time and in the theory book of Garratt (1994) they are still regarded as overall valid for moderate ranges of the ratio z/L (-5 < z/L < 1). Furthermore the stability functions using these estimations of  $\alpha$  and  $\gamma$  serve as input in the parameterisation of the aerodynamic resistance in the EMEP (European Monitoring and Evaluation Programm) deposition module (Simpson et al., 2002). Depending on the site properties, such as vegetation type or geographical latitude, and the available data, other observations suggest slightly different values for the two parameters. According to the summary of Garratt (1994)  $\alpha$  is found to be between 4.7 and 9.2 and the reported values of  $\gamma$  lie in the range of 9 to 16. Here the recommendations of van Ulden and Holtslag (1985) are used in order to be consistent with the calculations of Hole et al. (2004) who reported ozone fluxes at the same site in Hurdal with earlier data just from the weather station. To understand the importance of the constants in the stability functions for the results, the flux calculations have also been performed setting  $\alpha$ to 8 and  $\gamma$  to 12 as done by Högström (1988). This procedure additionally enables an improved view on the differences in the flux calculations resulting of the choice of the stability function. Mikkelsen et al. (2000) used the same values in their performance, thus a direct comparison between the two parameterisations independent of the constants is allowed.

To get a better idea of the data quality provided by the different stations and as another reference, the flux has also been calculated replacing the friction velocity by the wind gradient measurements leading to

$$F = -\frac{k^2(u(z_2) - u(z_2))(c(z_2) - c(z_1))}{\left(\ln\frac{z_2 - d}{z_1 - d} + \psi_h((z_1 - d)/L) - \psi_h((z_2 - d)/L)\right)^2}$$
(9)

where u(z) denotes the wind speed at the height z. This equation represents the original formula used by Mikkelsen et al. (2000) when no sonic data were available.

In addition F has been estimated using a transfer coefficient directly obtained from the sonic measurements according to Mikkelsen et al. (2004). The factor is simply given by

$$f = \frac{u_*^2}{u(z_2) - u(z_1)} \tag{10}$$

and also appears in other flux studies in literature where sonic measurements were performed (e.g. Pilegaard et al., 1995).

#### 4.2 Roughness length

Based on the logarithmic wind law for neutral conditions the aerodynamic roughness length  $z_0$  (i.e. the height where the wind speed becomes zero) was calculated using the equation

$$z_0 = \frac{z - d}{\exp\left(\frac{ku(z)}{u_*}\right)} \qquad \text{for } 0 \le H < 10 \text{ [Wm^{-2}]}$$
(11)

where z was set to the upper measurement height (25 m). The limits for the sensible heat flux constrain the dataset to only near-neutral conditions and hence allow to approach  $z_0$  by Eq. (11). Averaged over the three measurement years a roughness length of 1.5 m was estimated for the Hurdal spruce forest, varying from 1.34 m in 2004 to 1.47 m in 2005 and 1.69 m in 2006. Similar length scales were found for a northern boreal forest in Finland with the same canopy height as in Hurdal. There the calculated values for  $z_0$  were in the range between 1 m and 1.9 m depending on the wind direction (Rinne et al., 2000). An average length of 1.75 m independent of wind speed and direction was estimated over a mixed spruce-pine stand with a maximum height of 24.5 m in Sweden by Mölder et al. (1999). Graefe (2004) reviews surface parameters retrieved from different datasets in literature and finds values for  $z_0$  between 0.35 m and 1.15 m for the roughness length of forests in general, and a value of 1.1 m for a spruce forest with a canopy height of 12 m. To validate the results an alternative calculation of the roughness length given by Rinne et al. (2000) was performed using the constraint dataset that resulted in nearly similar values for all three years. However the missing sonic data is thought to influence the results, for the friction velocity is not reported during most of the winter months. This lack of small roughness lengths resulting out of winterly snow cover in the calculation of the annual average could explain the higher values compared to a roughness length of 0.5 m predicted by Hole et al. (2004) for the same site. Moreover it is assumed that the distinct data availability in the three years (see Figure 1) contributes to the differences in the average annual values. The same order of  $z_0$  in all three years and the similar pattern of its distribution (Figure 2) indicate that the meteorological and surface conditions did not change substantially from one year to another.

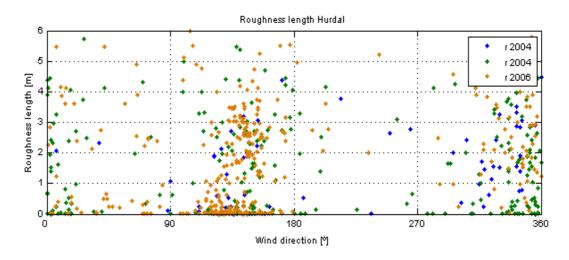


Figure 2: Roughness length against wind direction in different years.

No clear connection between the wind direction and the roughness length could be identified (Figure 2). This gives confidence that all measurements can be used for the gradient calculations and no further partition of the data according to the wind sector was carried out.

#### 4.3 Enhancement factor

Near very rough surfaces, such as forest canopies, the flux gradient laws tend to break down and flux-profile relationships have to be adapted to the region of the surface layer immediately above the canopy, the so-called ,roughness sublayer' (Cellier and Brunet, 1992; Simpson et al., 1998; Rinne et al., 2000). Comparisons between measured fluxes and those predicted by similarity theory show that the calculated values underestimate the transfer coefficients in the roughness sublayer (e.g. Thom et al., 1975; Högström et al., 1988; Simpson et al., 1998) and suggest, to correct the fluxes obtained by Eq. (1) by multiplying them by an enhancement factor

$$\gamma = \frac{\phi}{\phi^*} \tag{12}$$

where  $\phi$  is the dimensionless gradient of a scalar according to the Monin-Obukhov similarity theory and  $\phi^*$  is that according to measurements. Different  $\gamma$ -coefficients are observed depending on the measurement height and type of forest and discussed later on. In this report a mean enhancement factor,  $\Gamma$ , is used to correct the originally calculated fluxes. Following the recommendations of Rinne et al. (2000),  $\Gamma$  is obtained by integrating the  $\gamma$ -coefficient between the measurement heights  $z_1$  and  $z_2$ :

$$\Gamma(z_1, z_2) = \frac{1}{z_2 - z_1} \left[ \int_{z_1}^{z_1} \gamma(z) \, dz + \int_{z_*}^{z_*} dz \right] \qquad z_1 < z_* < z_2 \qquad (13)$$

The correction function is given by Cellier and Brunet (1992) and supported by the study by Mölder et al. (1999):

$$\gamma(z) = \frac{z_* - d}{z - d} \tag{14}$$

Here as well as in Eq. (13) d is the earlier introduced displacement height (8 m)and  $z_*$  is the roughness layer depth. The estimation of  $z_*$  is associated with uncertainties (Graefe, 2004), and conventionally two times the canopy height is used as a first approximation, i.e.  $z_* \approx 2h$  (Cellier and Brunet, 1992; Simpson et al., 1998). Several authors (Garrat, 1980; Cellier and Brunet, 1992) propose that z\* should be related to a length scale,  $\delta$ , characterizing the main source of horizontal inhomogenities of the canopy by  $(z_* - d)/\delta \approx 3 - 4$ . In case of forests,  $\delta$  is identical to the average horizontal spacing between the trees of the stand and was here approximated by forest inventory data of the Hurdal site (Andreassen et al., 2005). In the year 2004 the reported stem number was about 680 per ha and the average diameter of the spruces 260 mm. This leads to  $\delta \approx 4$  and therefore to a roughness sublaver depth of  $z_* \approx 24$  m, using  $z_* \approx 4 \delta + d$  (Rinne et al., 2000). The calculated length is in the order of two times the canopy height (26 m) and lies in the domain of Eq. (13), since  $z_*$  has to be less than  $z_2$  (25 m). Using the measurement heights of 15 m and 25 m and  $z_* = 24$  m, the  $\Gamma$ -coefficient to correct the Hurdal flux calculations was estimated to 1.42. This result is supported by measurements of Simpson et al. (1998) above a mixed forest who obtained  $\gamma$ -coefficients ranged between 1.1 and 1.8 for measurement height to canopy height ratios (z/h) between 2.2 and 1.2. Even higher  $\gamma$ -values are reported by Cellier and Brunet (1997) who review measurements above different canopies: The presented studies have measured transport coefficients above pine forests enhanced by factors of 1.3 up to 3 above those predicted by similarity theory.

#### **5** Results

#### 5.1 Annual and diurnal cycles of the measured data

Analysis of all directly measured parameters with relevance for the flux calculations will be presented in this section as an overview of the conditions at the Hurdal site, as a first approach to the predominant flux regime and in order to estimate the quality of the measurements. The results are reported in terms of annual and diurnal cycles that are based on monthly mean values respectively hourly mean values (Figures, see Appendix).

In Figure 3 annual temperature cycles at the upper measurement height (25 m) are presented, showing the continental climate conditions in Hurdal with a temperature peak in July and lowest average temperatures in January and February. The average diurnal pattern of the variable is shown in Figure 4, splitted up in cycles for the four seasons winter (December – February), spring (March – May), summer (June – August), and fall (September – November). The daily temperature in all seasons is highest around 4 pm local time and characterized by smoother curves in fall and winter and higher gradients in the days of spring and summer. Strongly related to the air temperature is the sensible heat flux with an

annual peak of averagely 75 Wm<sup>-2</sup> earlier in summer (Figure 5). During winter months the flux becomes negative, indicating the cooling of the landmasses in continental climate conditions (by convention the flux is negative when examined from the atmosphere downwards to the ecosystem). The daily pattern (Figure 6) shows the largest sensible heat fluxes (up to 200 Wm<sup>-2</sup> in summer) at noon and explains the above described temperature peaks taking into account a lag time to warm the air. Diurnal sensible heat fluxes measured for single summer days above a spruce-pine forest in Uppsala by Mölder et al. (1999) show good agreement in timing of the cycles and size order of the values. Negative fluxes are measured during night in all seasons and the daily amplitude increases with the intensity of the solar radiation. The very small amplitude and predominantly negative values of the heat flux in winter illustrate again the geographical latitude and continental characteristics of the Hurdal site.

Even if the annual cycle of the friction velocity varies moderately from year to year with no obvious pattern except from an evident summer peak (Figure 7), the analysis of the data shows a clear diurnal cycle (Figure 8). The sonic station measures an enhanced surface stress in the early afternoon for all seasons except for winter where the stable stratification over longer periods is thought to impede increased wind velocities near surface. Very similar diurnal profiles are found by Keronen et al. (2003) for a Scots pine stand in Finland, supporting not only the pattern and the scale of the measured values, but also the fairly high average winter friction velocities that were not expected. Again the study of Mölder et al. (1999) is a good reference showing diurnal cycles for the turbulent velocity scale for two summer days in Finland with an analog pattern as in Hurdal. It has to be noted that the measured maxima for the friction velocity as well as for the sensible heat flux there were higher than the one presented in Figure 8 resp. Figure 6 which is not surprising when comparing a single day with averaged values over several months.

Figure 9 shows the annual development of the ozone gradient, calculated from the measured concentrations at 25 m and 15 m in the three measurement years and as an overall average. Mean values of certain months vary fairly from year to year but in the cold season normally lower gradients are found and a minimum in June is assessed in all three years. An explanation of these characteristics and additional information is provided by a diurnal analysis of the parameter as seen in Figure 10.  $O_3$  gradients are highest in summer nights with measurements above  $8 \,\mu \text{gm}^{-3}$  and reach minimum values of around 1.5  $\mu \text{gm}^{-3}$  during daytime in the two warmer seasons. The calculations show a weak diurnal cycle in winter revealing that the stable stratification of the atmosphere inhibits mixing processes and that ozone uptake is reduced to a minimum as a consequence of snow and rime cover. In summer and spring the gradient decreases rapidly after sunrise leading to a period with a nearly constant small value, while in fall a clear peak at noon can be detected and the decline of the ozone difference between the two measurement heights occurs much slower. Karlsson et al. (2006) present ozone gradients over a spruce forest in Sweden averaged over 93 days with complete diurnal measurements between April and September 2003. The results are given as relative differences in ozone concentrations between the lower and higher measurement height and show a similar pattern as the diurnal summer cycle in Hurdal but with relatively less increased values of delta  $O_3$  during the midday hours.

The annual cycle of the mean  $O_3$  concentration (i.e. the average value between the two measurement heights) is very similar in the three measurement years and shows an expected pattern with highest values of 80 µgm<sup>-3</sup> in spring (Figure 11). Calculating seasonal concentration averages and comparing them to values reported by Hole et al. (2004) for the years 2000 until 2003 for the same site shows a slight increase in the mean  $O_3$  concentration for all seasons in the here considered period (2004 until 2006) compared to the previous measurements. While the concentration remains constant during winter days, a clear diurnal pattern can be spotted in the other seasons decreasing in its specification from summer to spring and fall (Figure 12). The peak in the ozone concentration at around 4 pm was also found by Pilegaard et al. (1995) and by Mikkelsen et al. (2004) above a Danish Norway Spruce forest and by Karlsson et al. (2006) above a Swedish Norway Spruce forest. It is thought to be a consequence of efficient mixing of  $O_3$  from above during day and decreased biogenic tree activity compared to the earlier hours in the day.

#### 5.2 Ozone fluxes

#### 5.2.1 Calculations according to Mikkelsen et al. (2000)

To describe the ozone flux regime in Hurdal, average diurnal and annual cycles of the transfer coefficient, the ozone flux and the deposition velocity are presented, calculated as discussed in Chapter 4 and corrected by the estimated enhancement factor of 1.42.

The transfer coefficient, resulting from the calculation with the profile parameterisations of Mikkelsen et al. (2000), demonstrates a diurnal pattern for all seasons with highest values in the middle of the day (Figure 13). In winter the pattern shows lowest specification and the coefficient is slightly more elevated during night than in the three other seasons where the values decrease to 0.1 ms<sup>-1</sup>. The cycles are in good agreement with Mikkelsen et al. (2004) who present a diurnal pattern of a transfer coefficient calculated with the same parameterisations over a Danish Norway Spruce forest. There the value of the parameter increased from a nightly average of 0.12 ms<sup>-1</sup> to a maximum of 0.28 ms<sup>-1</sup> at noon. The more moderate range of the calculations performed by Mikkelsen et al. (2004) is thought to be a consequence of using an annual average instead of splitting up the data into four seasons. Furthermore the enhancement factor that elevated the values in our performance was not used to correct the flux measurements in Denmark. The seasonal behaviour of the parameter is fairly variable from year to year and as can be seen in Figure 14 most of the monthly averages in winter are only covered by data from one measurement year. However a peak of around 0.25 ms<sup>-1</sup> in June is reported in all three years as well as smaller coefficients during the cold seasons.

Retaining the profile parameterisations of Mikkelsen et al. (2000) leads to an annual ozone flux pattern as shown in Figure 15. Highest fluxes, reaching values of -0.4  $\mu$ gm<sup>-2</sup>s<sup>-1</sup> averagely, are reported in the middle of spring and later on in fall, whereas the data cover in the summarized cycle is again unsatisfying, especially

in the colder seasons, and the peak in April is a consequence of the exceptionally high measurements in 2005. The large year-to-year variation is also found by Mikkelsen et al. (2004) at the earlier introduced site in Denmark, where monthly mean ozone fluxes from 1996 to 2000 separated into night and day are calculated. Peaks vary in size from -0.4  $\mu$ gm<sup>-2</sup>s<sup>-1</sup> during night to -0.7  $\mu$ gm<sup>-2</sup>s<sup>-1</sup> during day and also in the month when they are measured (from April to September). The average diurnal cycle of the flux is not nearly as smooth as the corresponding curve of the transfer coefficient, but still shows an evident pattern with higher values during daytime in spring and in summer (Figure 16) and corresponds well with reported fluxes above coniferous forests in Scandinavia. Several authors found maximum values in between 8 am and 4 pm local time in spring and summer, such as Pilegaard et al. (1995) and Mikkelsen et al. (2004) over a spruce forest in Denmark, or Keronen et al. (2003) over a Finish Pine stand. While these studies present diurnal differences for the flux in the warmer seasons spanning a factor 3, the gradient in Hurdal is less distinct with night values between  $-0.2 \ \mu gm^{-2}s^{-1}$  in spring and -0.3  $\mu$ gm<sup>-2</sup>s<sup>-1</sup> in summer and peaks of -0.45  $\mu$ gm<sup>-2</sup>s<sup>-1</sup>. In winter the diurnal pattern is almost missing and the flux at a constantly small level, as can be expected under Norwegian continental conditions. The daily development of the parameter in fall is rather noisy with values fluctuating between -0.3  $\mu$ gm<sup>-2</sup>s<sup>-1</sup> and -0.45  $\mu$ gm<sup>-2</sup>s<sup>-1</sup> and no evident pattern. Possible explanations and a further discussion of these profiles are presented later on in Chapter 6.

Changing the displacement height to 10 m did not improve or change the pattern of the diurnal cycle of the ozone flux and implies that this parameter is of an inferior importance in the chosen profile calculation method. However, the displacement height is used to estimate the depth of the roughness sublayer and can therefore influence the value of the enhancement factor, as discussed later on.

Calculation of the deposition velocity according to Eq. (2) results in an annual profile of the parameter as shown in Figure 17. The pattern differs substantially from an annual cycle presented by Hole et al. (2004) for the same site and a period between 2000 and 2003. Especially the high values in fall exceed average deposition velocities measured over other Scandinavian forests (e.g. Mikkelsen et al., 2004). As the ozone deposition is related to processes that are stimulated by high temperatures (e.g. surface reactions) and high light intensities (e.g. stomatal conductance) (Mikkelsen et al., 2004), the highest velocities are expected in the summer months and during the midday hours. The calculated diurnal cycles of the parameter (Figure 18) do not show the anticipated maxima either and rather erratic patterns are found for all seasons. Particularly the summer profile with a very early morning peak of about 11 mms<sup>-1</sup> and small values around noon and the high velocities in fall during day- and night-time that even exceed the summer maximum are strange and not reported elsewhere. Hole et al. (2004) and Keronen et al. (2003) who both present diurnal cycles of the deposition velocity in different seasons found maxima of 7 mms<sup>-1</sup> around 9 to 10 am and values not higher than 1mms<sup>-1</sup> during nighttime for the summer months and corresponding less distinct cycles for the rest of the year. A closer look on the deposition velocities is presented in an overall discussion of the ozone regime in Hurdal in the next chapter.

#### 5.2.2 Comparison between different calculation methods

As described in chapter 5 the flux calculations have also been performed with alternative parametrisations of the stability functions. Figure 19 and Figure 20 show a comparison between the different calculation methods proposed by Mikkelsen et al. (2000), van Ulden and Holtslag (1985) and Högström (1988) for the seasonal diurnal cycle and the annual cycle of the transfer coefficient. Both the daily and the annual pattern of the different calculations correspond very well, especially in winter and nighttime when lower values of the parameter are reported. The estimated coefficient is always highest using the suggestions of van Ulden and Holtslag (1985) and the difference compared to the parameterisation of Mikkelsen et al. (2000) is most evident in the peaks. The curve resulting from Högströms calculation (1988) who uses the same equations as van Ulden and Holtslag (1985) with alternative values for  $\alpha$  and  $\gamma$  (see chapter 4) deviates not remarkably from the most distinctive curve of van Ulden and Holstlag (1985). This indicates that the choice of the parameterisation has a slightly bigger influence on the results than the setting of the constants. However in general the profiles are found to be very similar also for the calculated ozone flux and the deposition velocity.

Replacing the friction velocity by the wind gradient measurements and calculating the flux parameters according to Eq. (9) resulted in a highly variable pattern. Since the plots could not provide additional information nor improve the results they are not shown here.

In Figure 21 a comparison of the transfer coefficient determined with the profile assumptions according to Mikkelsen et al. (2000) or obtained with sonic data (Eq. (10)) is shown. Both parameters have been corrected by the enhancement factor. The profile of the sonic coefficient is generally more variable, especially in the colder seasons. As a consequence of snow and ice cover the wind sensors measure more unreasonable values that have to be removed so that the data cover is poorer during these months. In winter and fall the sonic coefficient overestimates the one calculated by the profile method during the whole day, but shows a similar development. In the warmer seasons the sonic cycles are smoother with lower values for the coefficient during the day and higher ones during night-time. Mikkelsen et al. (2004) present an analog comparison between a sonic and a profile transfer coefficient over a Danish Norway Spruce canopy from continuous measurements during one year. As they did not distinguish between different seasons, Figure 22 shows a diurnal cycle for the two coefficients in Hurdal averaged over three measurement years to allow a better comparison with the reference. The summarized diurnal patterns of the two coefficients in Hurdal correspond not as well as they did in the calculations of Mikkelsen et al. (2000), but still have a related development. Contrary to the measurements in Denmark that picture an overall underestimation of the sonic coefficient during night-time and an overestimation in the peak zone, the situation in Hurdal is found to be reverse as it was described before for the warmer seasons. Mikkelsen et al. (2000) detected the biggest difference between the two measurements during day in May and during night in September. As Figure 21 illustrates the results obtained for the Hurdal site support these larger deviations around noon in Spring and at night-time in Fall. However the fact that contrary characteristics of the profile and sonic cycles were found for the two measurement sites remains.

#### **6** Discussion

#### 6.1 Enhancement factor

The flux calculations in Hurdal were corrected by an enhancement factor estimated from variables that can take different values depending on the measurement settings and the approximation method. In order to evaluate the influence of the measurement heights and the depth of the roughness sublayer on the enhancement factor,  $\Gamma$  was also calculated using reasonable alternative values for  $z_1$ ,  $z_2$  and  $z_*$  (Table 1).

z <sub>1</sub> [m]	z <sub>2</sub> [m]	<i>z</i> ∗ [m]	Γ
15	25	24	1.42
15	30	24	1.28
15	50	24	1.12
20	35	24	1.04
15	50	39	1.63
20	50	39	1.34

 Table 1:
 Enhancement factor calculated from different length scales.

The upper part of Table 1 illustrates the importance of the measurement levels. Given the calculated roughness sublayer height of 24 m, the enhancement factor decreases with an increasing upper measurement height and can be minimized by elevate the lower height close to z\*. Cellier and Brunet (1992) present similar results and show that the errors made when applying the classical aerodynamic method to profiles recorded in the roughness layer increase rapidly when  $z_*$ ,moves' upwards in the region  $z_1 < z_* < z_2$ . Changing the roughness layer depth (e.g. when varying the displacement height) supports the conclusion that measurement heights should be chosen according to  $z_* - z_1 \le z_2 - z_*$  if similarity theory is used to calculate fluxes. Simpson et al. (1998) state that deteriorated enhancement factors above 1.6 reduce the reasonability of chosing the fluxgradient method to calculate deposition fluxes. Therefore they suggest to set the sampling stations at heights of at least 1.4 the canopy height (i.e.  $z/h \ge 1.4$ ). Practically there are of course some limitations to this recommendation, e.g. equipment becomes more expensive, sensors become less accessible and the measured gradients smaller (Cellier and Brunet, 1992). Although the inclusion of the correction functions in the calculation helps to improve the results substantially (Mölder et al., 1999) and in awareness of the practical constraints, it would clearly improve the quality of the measurements if the upper measurement height in Hurdal was elevated by using a bigger tower. This recommendation is supported by the fact that the estimation of  $z_*$  is uncertain and other authors present significantly higher values for the roughness sublayer depth above forests than the one calculated here (Garrat, 1994; Mölder et al., 1999).

#### 6.2 Annual and diurnal ozone flux in Hurdal

In order to report the ozone flux regime in Hurdal, annual and diurnal cycles of the transfer coefficient, the ozone flux and the deposition velocity based on data series of three years have been presented. While the pattern of the transfer coefficient is in good agreement with previous studies (Mikkelsen et al., 2000; Mikkelsen et al., 2004), the resulting deposition fluxes and velocities show rather noisy and partly unexpected diurnal and annual profiles. Still a few common characteristics of fluxes over Scandinavian forests are also assessed at the Hurdal site.

The clear difference between summer and winter in the cycles and a very weak diurnal pattern in the colder season represent the Norwegian continental conditions and were also found in other comparable flux studies (e.g. Hole et al., 2004; Mikkelsen et al., 2004). Longer periods of elevated ozone flux in summer than in the rest of the year (Figure 16) reflect the importance of the distinctively different seasonal light and temperature conditions (see also Figure 4 and Figure 6) for the deposition processes and are also reported by Hole et al. (2004) for the Hurdal site and by Keronen et al. (2003) over a Finish pine stand.

More difficult to explain are the presented profiles in fall which are not only very noisy but also exceed in their values the calculations of Hole et al. (2004) substantially. One possible reason for the highly variable patterns is the poor data availability in the fall and winter months. As mentioned in Chapter 3 (Figure 1) the lacking sonic measurements, as well as the combination of data from different stations, lead to high percentages of missing values of the ozone flux and the deposition velocity. Thus average values are based on few measurements and become less meaningful. Smoother curves could then be expected if more data were available as seen when plotting the annual diurnal cycle of the transfer coefficient averaged over a whole year compared to the seasonal profiles (Figure 21 and Figure 22). Practically it is difficult to avoid the data lacks during the cold and snowy months, thus it is recommended, if measurements are carried out, to consider the whole data series starting in 2003 in future calculations, especially of the critical parameters.

Keronen et. al. (2003) report higher deposition velocities in October than in March for the year 2002 over a Finish pine forest and support the here presented result that still considerable deposition processes can occur in fall (Figure 17 and Figure 18). Depending on the meteorological conditions the physiological activities of the vegetation in a particular month vary from year to year and influence not only the annual but also the diurnal average profiles of the flux parameters (Keronen et al., 2003). The reported high deposition velocities in the October of 2005 (Figure 17) are considered as exceptions and together with the poor database thus partly explain the elevated diurnal cycle of the parameter in fall. However, the situation pictured by Hole et al. (2004) with significantly smaller deposition velocities in fall than in spring in the annual, as well as in the diurnal pattern, is thought to be much more representative for the Hurdal site than the plots presented here.

The calculated fluxes and deposition velocities were relatively high during night (Figure 16 and Figure 18) with values of more than half the daytime deposition,

especially in summer. This indicates that ozone removal processes other than stomatal uptake may contribute significantly to the observed flux, such as reaction with plant surfaces, particles and reactive gases emitted from soil (Pilegaard et al., 1995). By contrast Hole et al. (2004) reported much smaller deposition velocities during night between 2000 and 2003 in Hurdal and thus assumed that the nonstomatal fraction is fairly small. Taking into account the cold climate in the region studied, their results are again much more reasonable than the ones obtained in this report.

Figure 18The peak of the summer deposition velocity in earlier morning hours as shown in Figure 18 is also found by other authors albeit not as soon after sunrise. Mikkelsen et al. (2000) present highest velocities over a Danish Norway spruce forest at 8 am local time, as well as Keronen at. al (2003) over a Scots pine stand in Finland. However, the fast decrease of the curve after the maximum and the low values in the middle of the day and in the afternoon are difficult to understand and remain unexplained.

#### 6.3 Different calculation methods

Despite estimating ozone fluxes in Hurdal another aim of this report was to evaluate the influence of alternative calculation methods on the results. The annual and diurnal patterns for the different flux parameters were found to be very similar using the gradient method with alternative profile parameterisations (Figure 19 and Figure 20). Neither the choice of the stability function nor the setting of the widely discussed constants  $\alpha$  and  $\gamma$  seemed to change the cycles substantially.

A slightly stronger influence could be detected when using a sonic transfer coefficient without any profile assumptions (Figure 21 and Figure 22). Still the two approaches led to similar patterns for the deposition velocity with the same unexpected characteristics as discussed before. This indicates that the cause of the variable profiles does not lie in the calculation method but is rather thought to be related with the data base and thus with the measurements. This conclusion is supported by other studies in the literature that compare different calculation methods and usually find similar flux patterns. Keronen et al. (2003) as well as Mikkelsen et al. (2000) found generally good agreements between the eddy covariance method and the gradient method with some smaller discrepancies in the morning hours. The latter authors also provide a comparison of a profile and a sonic transfer coefficient similar to the one presented in this report with corresponding diurnal cycles (Mikkelsen et al., 2004).

The recently installed sonic station in Hurdal improved the results compared to calculations using only profile parameters, even if the data cover was very poor during the winter months. The ice cover on the sensors is a common problem when using anemometers in cold Scandinavian conditions and can lead to longer periods of inoperative measurement stations (Keronen et al., 2003), as it was the case in Hurdal (Figure 1).

From this study it appears that improvements in the measurement settings at the Hurdal site could help to provide better data to calculate ozone fluxes with the described profile method. Annual and diurnal cycles of directly measured meteorological parameters and ozone concentrations and gradient show wellknown patterns and represent the continental characteristics of the site. Still rather variable and partly unexpected cycles for characteristic flux parameters resulted from calculations by the gradient method and reliable conclusions are difficult to draw. Using different profile parameterisations and alternative transfer coefficients did not change the results substantially and indicated that the poor quality of the calculations is caused by other constraints. A discussion of the Enhancement factor to correct flux calculations over forest canopies showed that elevating the upper measurement height by at least 15 m could make the results more reliable and representative. Another source of the erratic cycles was the fairly poor data base when calculating mean values resulting from the malfunction of the two measurement stations during different periods and nearly no available data from the sonic station in winter months. Since the icing of the sensors is practically hard to avoid it is suggested to consider longer time series when calculating seasonal diurnal cycles to reduce the influence of exceptional measurements and the high year to year variability.

#### 7 Acknowledgements

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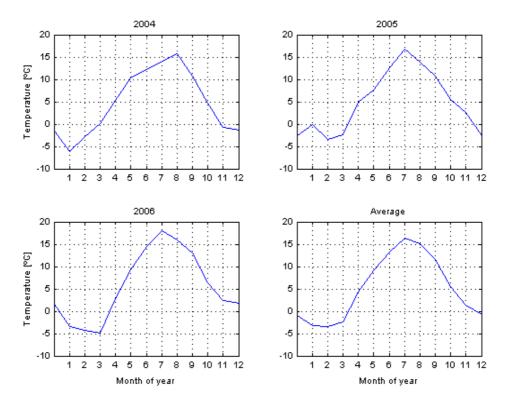
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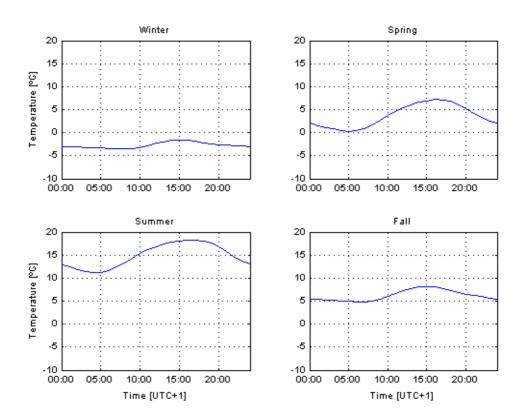
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Appendix A

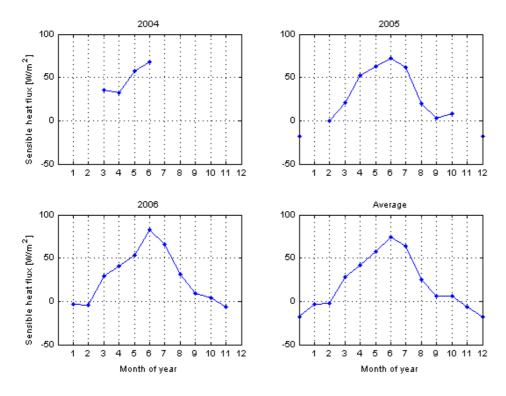
Figures 3–22



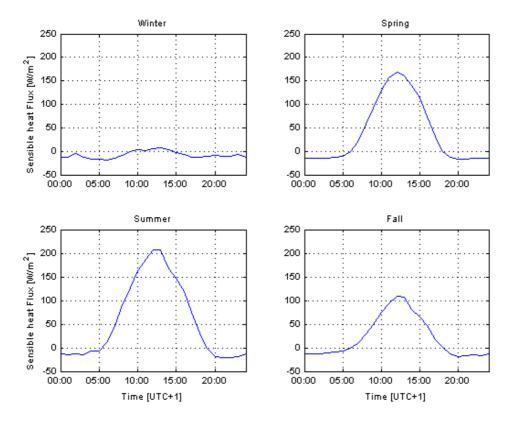
*Figure 3:* Average annual cycles of the temperature at 25 m in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years.



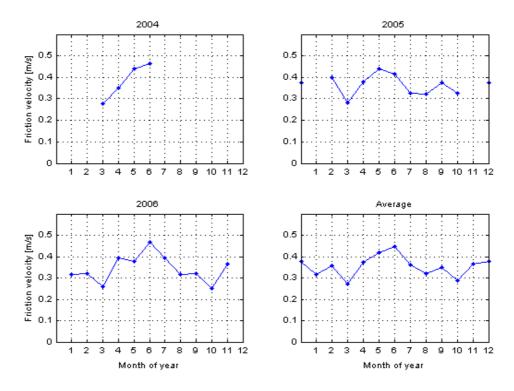
*Figure 4:* Average diurnal cycles of the temperature at 25 m in Hurdal for winter, summer, spring and fall.



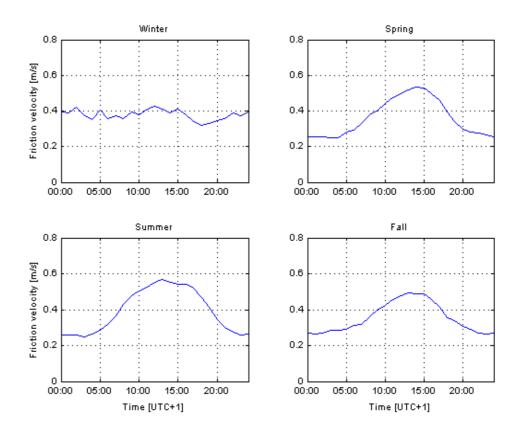
*Figure 5: Average annual cycles of the sensible heat flux in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years.* 



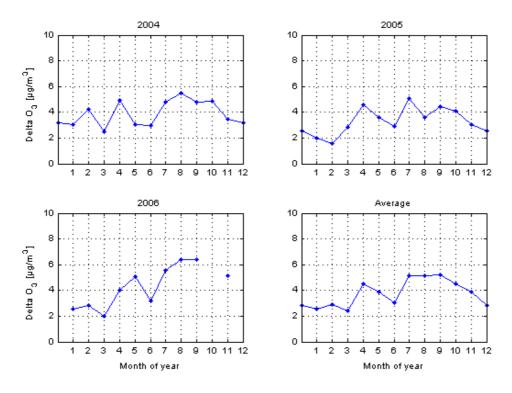
*Figure 6:* Average diurnal cycles of the sensible heat flux in Hurdal for winter, summer, spring and fall.



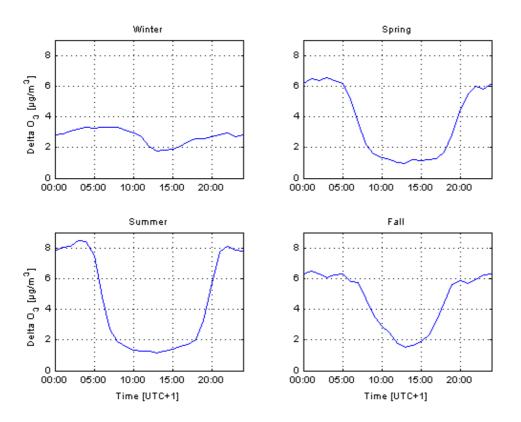
*Figure 7: Average annual cycles of the friction velocity in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years.* 



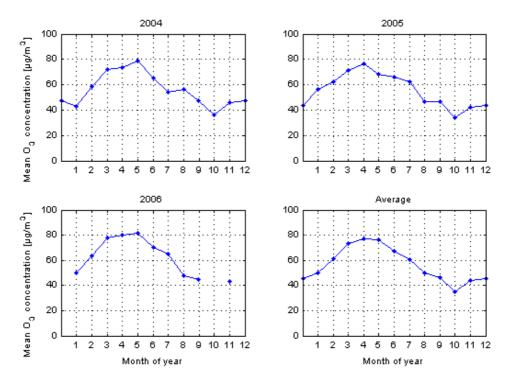
*Figure 8: Average diurnal cycles of the friction velocity in Hurdal for winter, summer, spring and fall.* 



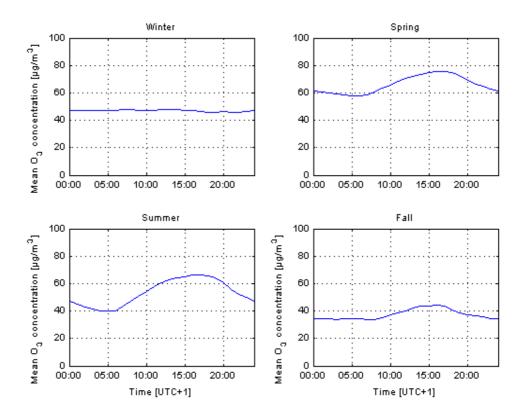
*Figure 9:* Average annual cycles of the concentration difference of ozone between 25 m and 15 m in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years.



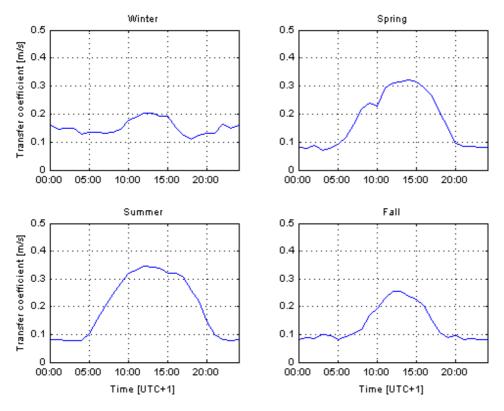
*Figure 10: Average diurnal cycles of the concentration difference of ozone between 25 m and 15 m in Hurdal for winter, summer, spring and fall.* 



*Figure 11: Average annual cycles of the mean ozone concentration in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years.* 



*Figure 12: Average diurnal cycles of the mean ozone concentration in Hurdal for winter, summer, spring and fall.* 



*Figure 13: Average diurnal cycles of the transfer coefficient in Hurdal for winter, summer, spring and fall, calculated according to Mikkelsen et al. (2000).* 

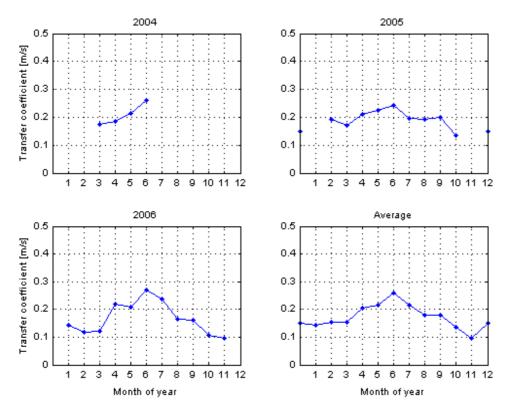
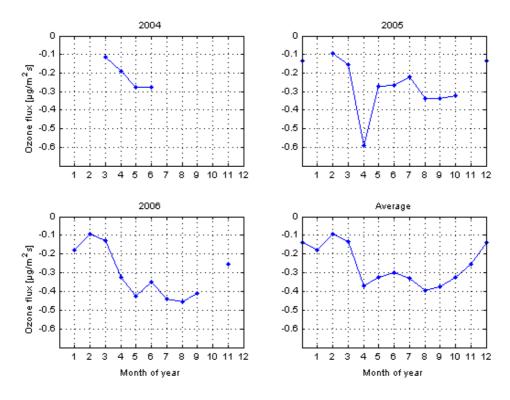
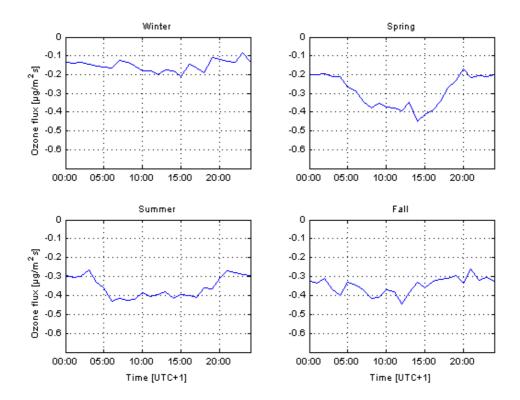


Figure 14: Average annual cycles of the transfer coefficient in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years, calculated according to Mikkelsen et al. (2000).



*Figure 15: Average annual cycles of the ozone flux in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years, calculated according to Mikkelsen et al. (2000).* 



*Figure 16: Average diurnal cycles of the ozone flux in Hurdal for winter, summer, spring and fall, calculated according to Mikkelsen et al.* (2000).

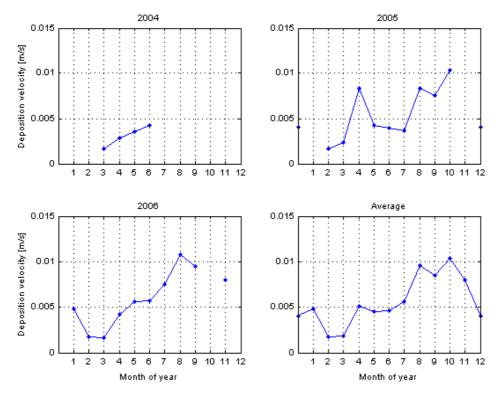
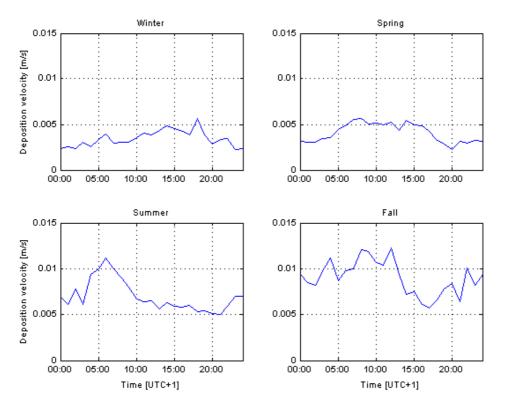


Figure 17: Average annual cycles of the deposition velocity in Hurdal in 2004, 2005 and 2006 and averaged over the three measurement years, calculated according to Mikkelsen et al. (2000).



*Figure 18: Average diurnal cycles of the deposition velocity in Hurdal for winter, summer, spring and fall, calculated according to Mikkelsen et al. (2000).* 

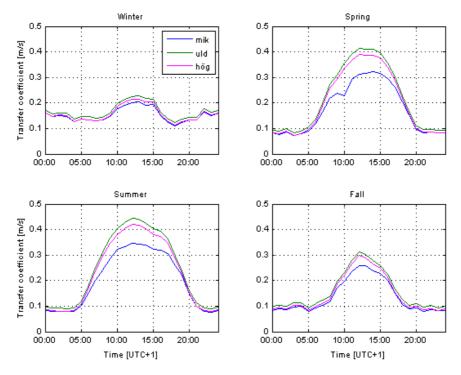


Figure 19: Average diurnal cycles of the transfer coefficient in Hurdal calculated with different parameterisations of the stability functions. Mik = Mikkelsen et al. (2000), uld = van Ulden and Holtslag (1985), hög = Högström (1988).

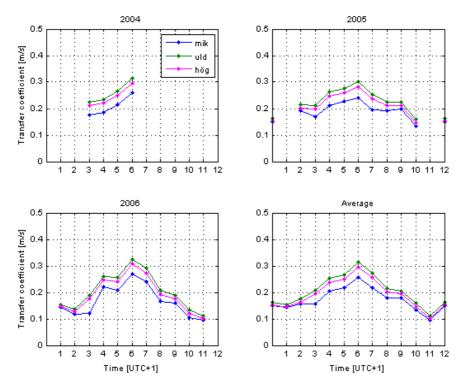


Figure 20: Average annual cycles of the transfer coefficient in Hurdal calculated with different parameterisations of the stability functions. Mik = Mikkelsen et al. (2000), uld = van Ulden and Holtslag (1985), hög = Högström (1988).

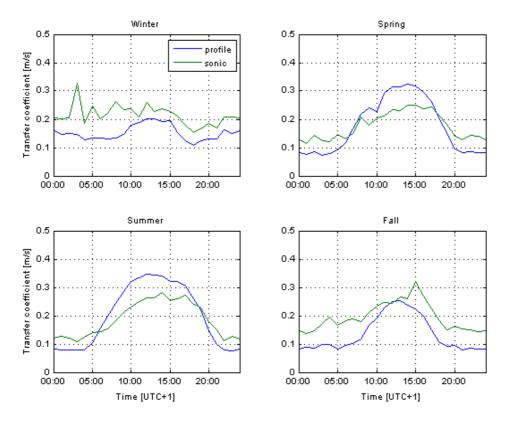


Figure 21: Average diurnal cycles of the transfer coefficient in Hurdal for the different seasons calculated from profile assumptions according to Mikkelsen et al. (2000) or determined with sonic data.

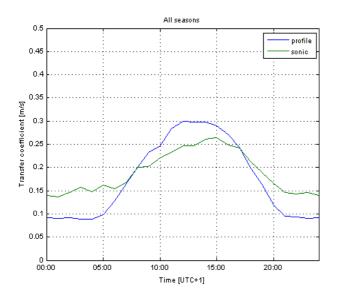


Figure 22: Average diurnal cycle of the transfer coefficient in Hurdal over the measurement years calculated from profile assumptions according to Mikkelsen et al. (2000) or determined with sonic data.



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Estimates of diurnal and seasonal variation of ozone flux parameters to a temperate coniferous forest over 3 years in Southern Norway are presented. The turbulent transfer coefficient, the ozone flux and the deposition velocity are calculated by the gradient method using alternative profile parameterisations. Even if annual and diurnal profiles of directly measured meteorological and sonic parameters show well known patterns, the corresponding resulting curves for the ozone flux and the deposition velocity are highly variable with unexpected elevated values during night, especially in fall. Performances with alternative profile parameterisations and a comparison between the turbulent and a sonic transfer coefficient correspond very well, particularly in the timing of the profiles. Improved results are expected if longer time series are considered in the calculations of the daily and monthly averages. Additionally the elevation of the upper measurement height is thought to ameliorate the measurements and data base.						
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