

2 Trends in positive and negative ozone laminae 3 in the Northern Hemisphere

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7 [1] The measured ozone profile is often not a smooth curve with a maximum in the
8 stratosphere. It exhibits narrow layers of enhanced ozone concentration (positive laminae)
9 and of depleted ozone (negative laminae). Here we deal with the trends in ozone laminae
10 characteristics. All sufficiently long data series of ozonesonde soundings from the
11 Northern Hemisphere poleward of 30°N are analyzed separately for Europe, northern
12 America, Japan, and the Arctic. The trends in ozone laminae are quite strong, much
13 stronger than those in total ozone at middle latitudes. A reversal in trends in the
14 ozone laminae characteristics, mainly in the overall ozone content (deficit) in positive
15 (negative) laminae per profile and the number of laminae per profile, is found to have
16 occurred in the mid-1990s. Whereas a negative trend was observed before the mid-1990s,
17 a positive trend was observed after about 1995. We assume this change in the ozone
18 laminae trend to be caused predominantly by a change in the trends in circulation in the
19 middle atmosphere.

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23 1. Introduction

24 [2] The ozone profiles measured by ozonesondes, particu-
25 larly those observed in late winter and early spring, do not
26 display a smooth shape below the maximum of the ozone
27 layer. Very often we may observe the occurrence of weak
28 undulations and/or relatively narrow layers of substantially
29 increased or depleted ozone concentration. These layers are
30 called laminae, positive laminae in the former (enhanced
31 ozone concentration) and negative laminae in the latter case.
32 The laminar structure of ozone profiles was first described
33 by *Dobson* [1973] on the basis of ozonesonde data. Lam-
34 inae occur also in lidar and satellite ozone profiles [e.g.,
35 *Appenzeller and Holton*, 1997; *Manney et al.*, 2000, 2001;
36 *Randall et al.*, 2003] but with poorer height resolution
37 (satellites about 1 km at best) and with much shorter data
38 series than ozonesonde data available at middle latitudes
39 since the 1960s or early 1970s. We are interested in long-
40 term trends and therefore hereafter we deal with ozonesonde
41 data only. On the other hand, satellites provide global
42 coverage and more frequent measurements (sondes typically
43 no more than 2–3 times per week at best). The first tracer
44 lamina climatology based on satellite data was published by
45 *Appenzeller and Holton* [1997]. However, that climatology
46 is based on calculations of where laminae are expected to
47 occur from horizontal gradients in the poor vertical resolu-
48 tion Microwave Limb Sounder data, rather than from direct
49 observations of laminae. *Kar et al.* [2002] used satellite data
50 from Stratospheric Aerosol and Gas Experiment (SAGE) II

(version 6.0). They were interested in layers with depths of 51
3 to 6 km, and they claimed that they had investigated 52
layers broader than laminae. 53

[3] Why do we study ozone laminae? First, in no other 54
parameter in the midlatitude middle atmosphere can we find 55
such a strong negative trend as in positive laminae over the 56
period of the late 1960s to early 1990s [e.g., *Lastovicka*, 57
2002]. Second, despite the small amount of ozone in the 58
individual laminae, the observed strong trends in laminae 59
could contribute to the observed trends in total ozone at 60
middle latitudes, particularly in late winter–early spring 61
[e.g., *Lastovicka*, 2001]. Third, the trends in laminae seem 62
to be, to some extent, related to long-term changes in 63
circulation/transport [e.g., *Mlch and Lastovicka*, 1997]. 64

[4] We deal with ozone number density profiles and 65
laminae expressed in terms of ozone partial pressure, not 66
with mixing ratio profiles. Strong laminae in terms of ozone 67
partial pressure occur only below the maximum of the 68
ozone profile, whereas strong laminae in terms of the ozone 69
mixing ratio also occur in the upper stratosphere. 70

[5] Laminae are observed most prominently during the 71
winter and spring in extratropics [*Reid and Vaughan*, 1991]. 72
At middle latitudes in Europe the seasonal variation of the 73
lamina occurrence frequency is larger than a factor of five 74
with a maximum in late winter–early spring [*Mlch and* 75
Lastovicka, 1996]. There are many profiles, particularly in 76
late summer and early autumn, which exhibit no stronger 77
lamina. On the other hand, in late winter and early spring 78
we can often observe profiles with more than one lamina, as 79
that shown in Figure 1. 80

[6] Laminae occur most frequently at heights around 81
14 km according to *Reid and Vaughan* [1991], which is 82

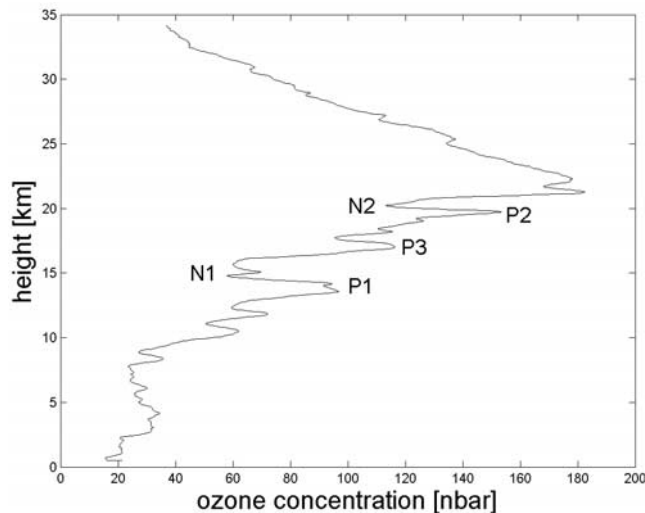


Figure 1. Ozone profile measured at Payerne on 11 February 2000. P1, P2, P3, positive laminations; N1, N2, negative laminations.

83 consistent with our result [Lastovicka, 2002] that more
84 than 40% of the overall ozone content in laminations is
85 located between 100 and 200 hPa. Other results [Bird *et al.*,
86 1997; Pierce and Grant, 1998; Orsolini *et al.*, 2001]
87 put the maximum occurrence of laminations at potential
88 temperatures of 375–400 K, i.e., about 14–16 km, which
89 agrees with the above results. Manney *et al.* [2000] found
90 for laminations in the lower stratosphere in November 1994
91 a pronounced longitudinal dependence with minimum
92 occurrence at 30–90°E and maximums at 90–150°E and
93 210–270°E.

94 [7] The laminations at high and middle latitudes are predom-
95 inantly associated with the exchange processes in the
96 vicinity of the vortex edge and therefore with the transport
97 of polar air in the form of oblique filaments toward the
98 midlatitudes [e.g., Mlch and Lastovicka, 1996; Reid *et al.*,
99 1994, 1998]. The filamentary structures are a part of tracer
100 (for instance ozone) sheets, vertically tilted in the shear zone
101 in the vicinity of the polar jet. Thin laminar structures
102 (positive or negative laminations) in the tracer vertical profiles
103 appear as the result of isentropic wrapping and vertical
104 shearing of such tracer sheets [Orsolini *et al.*, 1995;
105 Orsolini, 1995]. High-resolution modeling confirms that
106 tilted ozone sheets, peeled off near the vortex edge, result
107 in the formation of laminations in the ozone profile [Orsolini
108 *et al.*, 1997]. Balloon-borne measurements confirm well-
109 resolved laminations near the vortex edge [Orsolini *et al.*,
110 1998]. Manney *et al.* [2001] attributes multisatellite ob-
111 served laminations in early November 1994 to the protovortex.
112 At high latitudes inside the polar vortex there are two other
113 mechanisms of lamina formation, intrusions into the vortex,
114 and differential advection of local ozone anomalies (possi-
115 bly resulting from chemical loss) [Manney *et al.*, 1998]. The
116 poleward motion of filaments can create positive “super-
117 laminations” inside the Antarctic ozone hole as observed
118 by Moustouli *et al.* [2003]. Complex measurements at
119 Shigaraki (35°N) in April 1998 identified a well-developed
120 lamina caused by differential advection due to the vertical
121 shear associated with the subtropical westerly jet and the

stationary Rossby wave embedded therein [Tomikawa *et al.*, 122
2002]. Intrusions of ozone-poor tropical air into higher 123
latitudes can create negative laminations there [e.g., Manney 124
et al., 2000]. Some smaller undulations of the ozone profile 125
and weaker layers of enhanced or depleted ozone concen- 126
tration may be caused by gravity waves, particularly at 127
middle latitudes [e.g., Reid *et al.*, 1994; Pierce and Grant, 128
1998]. Therefore we consider only sufficiently strong lam- 129
inations (see section 2), which may be assumed to originate 130
very predominantly at the polar vortex edge. 131

[8] Since we are interested in trends in ozone laminations, 132
recent reviews on trends in related stratospheric parameters, 133
namely in the total ozone [Staehelin *et al.*, 2001] and 134
stratospheric temperatures [Ramaswamy *et al.*, 2001], 135
should be mentioned. Logan *et al.* [1999] have broadly 136
studied the trends in ozone profiles. 137

[9] Our previous investigations, based on the ozonesonde 138
data from the late 1960s to the early 1990s [Mlch and 139
Lastovicka, 1996, 1997; Lastovicka, 2001, 2002], yielded 140
the following main conclusions about laminations: 141

[10] 1. There is a very strong negative trend in the overall 142
ozone content in laminations per profile at middle and high 143
latitudes of the Northern Hemisphere. A typical decrease is 144
about 50% over 20–25 years. 145

[11] 2. This strong negative trend does not exhibit a 146
pronounced dependence on latitude down to about 36– 147
37°N (Tateno) despite the large decrease of the overall 148
ozone content in laminations per profile with decreasing latitude. 149

[12] There are various indications that long-term trends in 150
some parameters of the middle atmosphere at middle 151
latitudes of the Northern Hemisphere changed in the 152
1990s [Lastovicka and Krizan, 2005]. The change of trends 153
in laminations in the Northern Hemisphere is the main topic of 154
the paper. The change of trends in the positive laminations was 155
first mentioned by Krizan [2003] for Hohenpeissenberg and 156
Sodankylä in Europe. Such a change for the negative 157
laminations follows from the data presented by Tarasick *et al.* 158
[2004, Figure 3] for a few northern American stations. 159

[13] Section 2 deals with the lamina determination. 160
Section 3 briefly describes the data we have used. The 161
trends in both positive and negative laminations over Europe, 162
northern America, Japan and the Arctic are treated separately 163
for each region in section 4. The results are discussed in 164
section 5. Paper ends with section 6, conclusions. 165

2. Determination of Laminations

[14] Figure 1 shows an example of the measured ozone 167
vertical profile that clearly illustrates the main problem of 168
lamina calculation. It is the determination of a reference 169
profile, the ozone profile as it would be in the absence of 170
laminations, with respect to which the laminations are computed. 171
Various authors have used various approaches to the refer- 172
ence profile determination, which is one of the reasons for 173
some differences between their results. It should be noted 174
that ozone profiles, which are “wilder” than that in Figure 1, 175
are not rare. 176

[15] In this paper we use a method similar to that used by 177
Pierce and Grant [1998]. Its basic idea is to obtain the 178
reference profile by smoothing the observed profile. This 179
can be done by splines. We use a simpler 1-2-1 smoothing 180
applied repeatedly until the profile between the tropopause 181

182 and the ozone profile maximum (= region of strong lamina
183 occurrence) becomes uniform. The approach is as follows:
184 [16] There are n measured data points $a_0 \dots a_n$ along the
185 profile. The value of the smoothed i th point, pa_i , is ($i \neq 0$
186 and $i \neq n$):

$$pa_i = (a_{i-1} + 2 a_i + a_{i+1})/4 \quad (1)$$

187 The value of the first point, pa_0 :

$$pa_0 = (2a_0 + a_1)/3 \quad (2)$$

190 The value of the last point, pa_n :

$$pa_n = (2a_n + a_{n-1})/3 \quad (3)$$

192 This procedure is repeated until the reference profile
193 between the tropopause and ozone concentration maximum)
194 becomes uniform. Such a reference profile is shown in
195 Figure 1. Our method of smoothing reduces slightly the
196 ozone profile maximum and can create some problems at
197 both ends of the profile, but none of these three regions is
198 the lamina occurrence region, where the reference ozone
199 profile seems to be correct. On the other hand, no method
200 yields a reference profile, which is quite certainly correct.
201 Some degree of uncertainty always remains.

202 [17] Once we have the reference ozone profile, we can
203 compute the intersection points of this profile with the
204 measured ozone profile. There is a local extreme of the
205 observed profile between two consecutive intersection
206 points. If the extreme is a maximum, we have three points:
207 the first intersection point, the maximum and the second
208 intersection point. Designate $H1(2)$ the height of the first
209 (second) intersection point in meters, $O1(2)$ the ozone
210 concentration at the first (second) intersection point, and
211 $Omax$ the ozone concentration in the maximum. If these
212 three points match the following criteria, they form a
213 positive ozone lamina:

$$500 < (H2 - H1) < 3500 \quad (4)$$

$$Omax - 0.5(O1 + O2) > sl, sl - \text{minimum size of lamina} \quad (5)$$

$$Omax - O1 > sl/2, Omax - O2 > sl/2 \quad (6)$$

219 The two intersection points and the local minimum of ozone
220 concentration form in a similar way a negative ozone
221 lamina.

222 [18] Some smaller undulations of the ozone profile and
223 weaker layers of enhanced or depleted ozone concentration
224 may be caused by gravity waves, particularly at moderate
225 latitudes [e.g., *Reid et al.*, 1994; *Pierce and Grant*, 1998].
226 To exclude them, we consider only sufficiently strong
227 laminae, usually those larger than 40 nbar, i.e., $sl = 40$ nbar
228 in (5). When we applied this criterion to Figure 1, where no
229 extreme is larger than 40 nbar, we found no strong lamina
230 for this particular day.

231 [19] We calculate three parameters characterizing the
232 laminae in ozone profiles: (1) the occurrence frequency of

laminae per profile, (2) the ozone content (deficit) per
positive (negative) lamina, and (3) the overall ozone content
in positive laminae (or deficit in negative laminae) per
profile. The third parameter is analyzed in the paper more
broadly than the first two, because it is the most relevant for
estimating the contribution of trends in laminae to the total
ozone trends.

[20] Different methods of lamina determination and def-
inition have some impact on the results of lamina studies. In
previous investigations we used three different methods for
lamina calculation. Prior to the method described above two
other methods assuming all laminae to be positive (i.e., in
Figure 1 the positive laminae P1, P2 and P3 began in the
maximums of negative laminae N1 and N2) and the
observed profile as a reference profile, and different con-
straints of the type of equation (6). The older method used
before 2001 was described by *Halenka and Mlch* [1996],
and the second method by *Krizan and Lastovicka* [2004].

[21] *Krizan and Lastovicka* [2004] compared the results
obtained by those three methods for nearby stations Payerne
(Switzerland) and Hohenpeissenberg (southern Germany).
Even though the absolute values of the overall ozone
content in positive laminae per profile were substantially
different, the relative trends (in percentage) were very close
to each other. The strong trends appear to be quite robust
and method-independent. As for condition (4), lamina
thickness between 0.5 and 3.5 km, Figure 2 (computed
with the second method described by *Krizan and Lastovicka*
[2004]) reveals the independence of the trend on lamina
thickness in spite of a principal difference in the value of the
overall ozone content in laminae as a function of lamina
thickness. Figure 2 shows that thin laminae (upper limit of
thickness of 1.5 km) almost do not contribute to the overall
ozone content in strong laminae ($sl > 40$ nbar). The lower
limit of 0.5 km was selected with respect to the ozone
sounding vertical resolution of about 150 m [*Harris et al.*,
1998], but anyway very thin laminae do not play a role in
the overall ozone content in laminae, as Figure 2 indicates.
The trend might depend on the parameter sl (5); however,
Krizan and Lastovicka [2004] found that the trends for $sl =$
40 and 70 nbar are very similar, and *Lastovicka* [2002]
observed a similarity of trends for $sl = 40$ and 20 nbar. This
means that the trends are almost independent of the lower
limit of lamina size, sl .

3. Data

[22] As mentioned above, we use ozonesonde data with a
sufficiently long period of observations from four regions at
middle and high latitudes of the Northern Hemisphere over
the period 1970–2003: (1) European midlatitude stations,
Payerne (46.49°N, 6.57°E, 1970–2002), Hohenpeissen-
berg (47.8°N, 11.02°E, 1970–2002), Lindenberg
(52.21°N, 14.12°E, 1975–2003), Legionowo (52.4°N,
20.97°E, 1979–2003), Prague-Libus (50.02°N, 14.45°E,
1979–2003, only January–April), Uccle (50.8°N, 4.35°E,
1970–2001); (2) northern American stations, Goose Bay
(53.32°N, 60.3°W, 1970–2003), Edmonton (53.55°N,
114.01°W, 1973–2003), Churchill (58.75°N, 94.07°W,
1974–2003), Wallops Island (37.93°N, 75.48°W, 1970–
1998); (3) Japanese stations, Tateno (36.05°N, 140.1°E,
1970–2003), Sapporo (43.05°N, 141.33°E, 1970–2003)

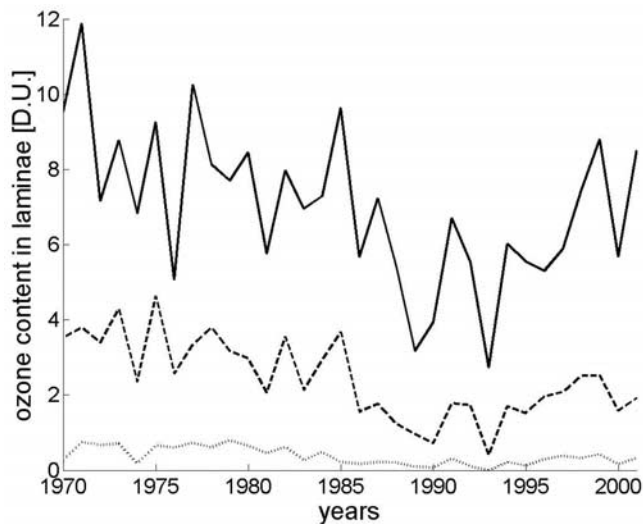


Figure 2. Overall ozone content in positive laminae per profile at Payerne in the period 1970–2001 for laminae larger than 40 nbar with the upper limit of lamina thickness 3.5 km (standard, top curve), 2.5 km (middle curve), and 1.5 km (bottom curve). After Krizan and Lastovicka [2004].

293 and Kagoshima (31.63°N, 130.6°E, 1970–2003); and
 294 (4) Arctic stations, Sodankylä (67.39°N, 26.65°E, 1989–
 295 2003), Ny Aalesund (78.93°N, 11.88°E, 1991–2003), Res-
 296 olute Bay (74.72°N, 94.98°W, 1970–2003), Alert (82.5°N,
 297 62.3°W, 1988–2003). Ozonesonde data for these stations
 298 were taken from the international ozone database in
 299 Toronto: <http://www.msc-smc.ec.gc.ca/woudc>. We use only
 300 data for January–May, the period of the highest occurrence
 301 frequency of laminae. In September–October, the number
 302 of laminae is less than 20% of that in February–April.

303 [23] There are large differences in the number of obser-
 304 vations at these stations. The following stations have the
 305 largest number of observations: Payerne, Hohenpeissen-
 306 berg, and Uccle. The Arctic stations except for Resolute
 307 Bay have shorter periods of observations. In the paper we
 308 report the results for stations of the Northern Hemisphere.
 309 The analysis of the ozone laminae characteristics in the
 310 Southern Hemisphere is under way and will be published in
 311 a separate paper.

312 [24] The intervals between data points in measured ozone
 313 profiles could vary from station to station and with time as
 314 measurements were improved. This can affect the results.
 315 The January–May averaged intervals between data points
 316 in profiles at “lamina” heights are summarized in Table 1
 317 for each station and each year. The smallest intervals,
 318 i.e., the best data, are provided by Payerne, Uccle and
 319 Sodankylä. The most recent data are best. Legionovo in
 320 1986 and Wallops till 1976 have average intervals larger
 321 than 1 km. However, Figure 2 shows that laminae thinner
 322 than 1.5 km contribute very little to the overall ozone
 323 content in laminae per profile for the laminae studied
 324 (>40 nbar). Therefore the varying intervals between data
 325 points in profiles probably do not affect significantly the
 326 results. This conclusion concerns particularly the long-term
 327 trends and it is supported by similarity of trend patterns at
 328 various stations, as shown in section 4. On the other hand,
 329 year-to-year variations may be affected to some extent.

[25] Another factor, the variable seasonal distribution of
 measurements, can affect the results due to a very strong
 seasonal variation of laminae. For example, Sofieva *et al.*
 [2004, Table 1] show distribution of monthly numbers of
 ozone sonde profiles for Sodankylä, 1989–1999. First,
 the yearly average number of profiles varies substantially,
 between 41 (1999) and 83 (1995), which may but need
 not affect the results. Second, and much more important,
 the seasonal distribution of soundings varies considerably
 from year to year, which can remarkably contribute to
 some large year-to-year changes observed at Sodankylä
 (see section 4, Figures 9 and 10). This is primarily
 problem of stations with smaller number of soundings.
 Stations with larger number and more regular soundings,
 like Payerne or Hohenpeissenberg, display smaller year-to-
 year changes (Figures 3 and 4).

[26] The third potential problem arises with differences in
 ozone sonde preparation and data processing between
 different countries, even different stations, and different
 types of sondes. This was a problem particularly in early
 years of measurements. Various intercomparisons, standard-
 ization, partial sonde unification, and data homogenization
 made this problem much less important in recent years
 [Harris *et al.*, 1998]. This may affect remarkably the
 differences in year-to-year variations among various sta-
 tions, particularly in the 1970s, but it does not seem to affect
 significantly the observed trends, because they are similar at
 various stations, as shown in section 4.

[27] The discussion in the two previous paragraphs shows
 that the gross features and strong long-term trends appear to
 be reliable, but fine details and year-to-year changes must
 be considered with great caution, and we do not consider
 many of them reliable.

4. Trends in Laminae

[28] Figures 3 and 4 display the trend in the overall ozone
 deficit (content) in negative (positive) laminae per profile
 for six European middle latitude stations. Figures 3 and 4
 look very busy and not very readable. However, this is to
 some extent our intention. Owing to the discussion at the
 end of section 3, only gross features, i.e., primarily the
 overall long-term trend of the whole data set, appear to be
 reliable, while year-to-year changes of individual curves are
 of very questionable reliability. We ask readers do not try to
 follow details, which might be incorrect and misleading,
 consider only the general character of long-term trends. The
 same is valid for all Figures 3–10.

[29] A principal change in the trend occurred in the mid-
 1990s. A strong negative trend (reduction of ozone content
 in positive laminae and reduction of ozone deficit in
 negative laminae) was observed before 1993–1995 (the
 minimum in 1993 might be the effect of the Mount Pinatubo
 volcanic eruption). In more recent years the trend is posi-
 tive, i.e., an increase of ozone content in positive laminae
 and ozone deficit in negative laminae. This change in trend
 in the overall ozone content in laminae per profile in the
 mid-1990s occurred both in the case of positive and
 negative laminae, and for all six individual stations. The
 negative trend in Figure 4 represents a laminar ozone
 content reduction by more than 50% from 1970 to the
 mid-1990s for Payerne, Hohenpeissenberg, and Uccle and a

t1.1 **Table 1.** January–May Average Values of the Interval Between Data Points in Profiles at Heights 10–25 km for All Stations and All Years^a

t1.2	Year	Pa	Hoh	Lind	Leg	Prg	Ucc	Sod	Ny	Res	Ale	Goos	Edm	Chur	Wal	Sap	Tat	Kag
t1.3	1970	165	385	0	0	0	133	0	0	536	0	609	0	0	954	684	749	865
t1.4	1971	171	381	0	0	0	139	0	0	609	0	620	0	0	1103	673	717	882
t1.5	1972	168	407	0	0	0	116	0	0	703	0	668	0	0	1247	688	702	864
t1.6	1973	177	438	0	0	0	120	0	0	630	0	671	591	0	1311	658	722	805
t1.7	1974	194	451	0	0	0	162	0	0	749	0	717	681	722	1111	646	572	831
t1.8	1975	184	427	636	0	0	176	0	0	853	0	805	769	815	1180	603	756	893
t1.9	1976	192	437	728	0	0	163	0	0	697	0	729	690	717	1138	698	1105	0
t1.10	1977	211	408	598	0	0	116	0	0	550	0	626	574	629	817	0	1246	0
t1.11	1978	228	409	670	0	0	128	0	0	633	0	648	598	628	793	553	824	759
t1.12	1979	218	366	666	765	619	138	0	0	609	0	646	580	655	637	609	642	748
t1.13	1980	232	345	664	882	681	140	0	0	610	0	623	545	603	551	710	752	1125
t1.14	1981	209	353	704	754	639	153	0	0	645	0	642	591	636	581	639	744	989
t1.15	1982	120	354	606	775	700	141	0	0	570	0	592	552	606	626	709	751	1041
t1.16	1983	116	367	555	793	701	115	0	0	581	0	587	577	582	0	678	734	1019
t1.17	1984	113	356	579	792	830	118	0	0	659	0	610	550	622	555	617	722	934
t1.18	1985	115	361	573	683	690	111	0	0	627	0	549	603	580	578	597	702	901
t1.19	1986	119	404	576	1069	689	107	0	0	573	0	577	550	543	577	673	687	867
t1.20	1987	135	384	645	723	770	120	0	0	400	0	399	367	377	522	664	720	851
t1.21	1988	137	386	781	857	545	112	0	0	368	342	386	361	366	537	714	732	909
t1.22	1989	139	384	867	777	631	120	52	0	354	330	367	349	336	594	678	726	886
t1.23	1990	28	404	742	651	605	83	50	387	405	373	401	395	407	556	673	744	891
t1.24	1991	25	379	661	633	633	89	63	128	310	296	370	320	317	524	651	710	901
t1.25	1992	25	385	602	678	536	86	303	79	322	332	381	325	356	560	676	714	957
t1.26	1993	21	385	560	262	508	84	53	293	433	406	391	379	392	560	707	797	928
t1.27	1994	21	359	437	188	474	84	50	52	345	382	275	267	322	524	690	768	897
t1.28	1995	20	367	466	62	489	79	51	54	287	298	260	301	337	561	687	733	878
t1.29	1996	22	427	453	54	508	74	46	52	250	320	314	311	301	527	655	753	858
t1.30	1997	21	350	408	52	499	77	12	54	270	40	279	355	294	494	625	700	820
t1.31	1998	22	330	438	52	467	82	12	59	251	263	296	316	279	501	559	678	750
t1.32	1999	20	359	423	53	454	82	11	61	262	260	306	316	261	0	599	721	816
t1.33	2000	21	342	450	49	53	85	10	62	36	40	48	41	48	0	407	417	545
t1.34	2001	34	356	440	50	53	89	11	58	39	40	49	37	46	0	378	447	533
t1.35	2002	35	342	467	50	54	83	9	55	43	41	47	39	45	0	415	454	522
t1.36	2003	32	356	438	48	54	83	9	55	41	41	45	39	42	0	57	58	62

^aIn meters. The 0 means no measurements. Pa, Payerne; Hoh, Hohenpeissenberg; Lind, Lindenberg; Leg, Legionovo; Prg, Prague-Libus; Ucc, Uccle; Sod, Sodankylä; Ny, Ny Aalesund; Res, Rolute Bay; Ale, Alert; Goos, Goose Bay; Edm, Edmonton; Chur, Churchill; Wal, Wallops Island; Sap, Sapporo; Tat, Tateno; Kag, Kagoshima.

t1.37

390 rapid trend reversal after the mid-1990s, Station Prague-
391 Libus provides larger values of both positive and negative
392 laminae because measurements were run only in January–
393 April each year, when the average number of laminae is
394 higher than in May. The comparison of Figures 3 and 4
395 reveals the ratio of the ozone deficit in negative laminae to
396 the ozone surplus in positive laminae to be about 2:3 or a
397 little lower with good correlation (but not one-to-one
398 correspondence) of the positive and negative laminae
399 variations.

400 [30] Canadian midlatitude stations (Figures 5 and 6)
401 display a trend pattern very similar to that for European
402 stations, a strong negative trend before the mid-1990s with a
403 minimum in 1995–1996 followed by a rapid trend reversal.
404 The Wallops Island data series terminated in the late 1990s.
405 Nevertheless, Wallops Island provides a similar trend pat-
406 tern for the positive laminae, whereas for the high-scatter
407 negative laminae data no evident trend can be detected. The
408 Wallops Island values of ozone in laminae are generally
409 somewhat smaller compared with Canadian stations due to
410 lower latitude. The ozone deficit to ozone surplus ratio
411 seems to be slightly smaller for the Canadian stations than
412 for the European stations.

413 [31] It is more difficult to draw a conclusion about trends
414 in the ozone content in laminae per profile for the Japanese
415 stations (Figures 7 and 8), because the number of observa-

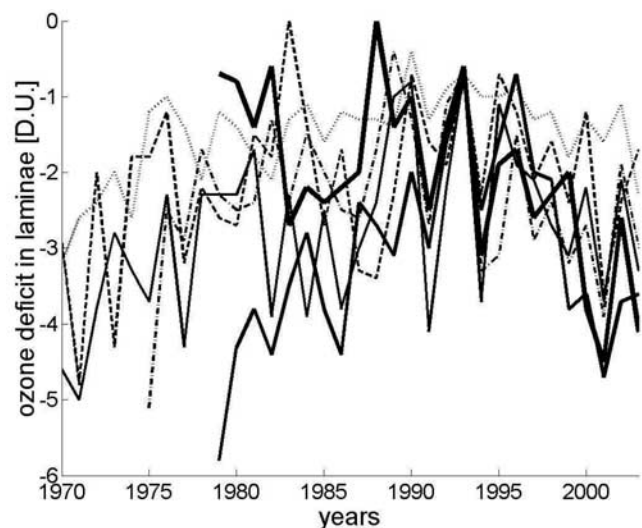


Figure 3. Overall ozone deficit in negative laminae per profile for the European middle latitude stations Hohenpeissenberg (dotted line), Legionovo (heavy solid line), Lindenberg (dash-dotted line), Praha-Libus (medium solid line), Payerne (thin solid line), and Uccle (dashed line), 1970–2003.

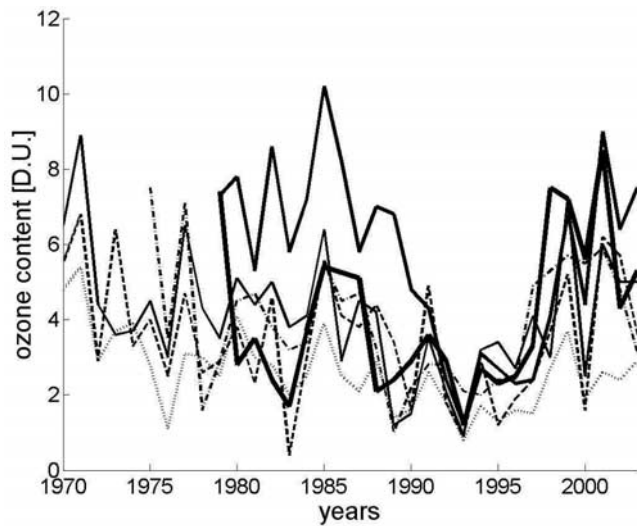


Figure 4. Overall ozone content in positive laminae per profile for the European middle latitude stations Hohenpeissenberg (dotted line), Legionovo (heavy solid line), Lindenber (dash-dotted line), Praha-Libus (medium solid line), Payerne (thin solid line), and Uccle (dashed line), 1970–2003.

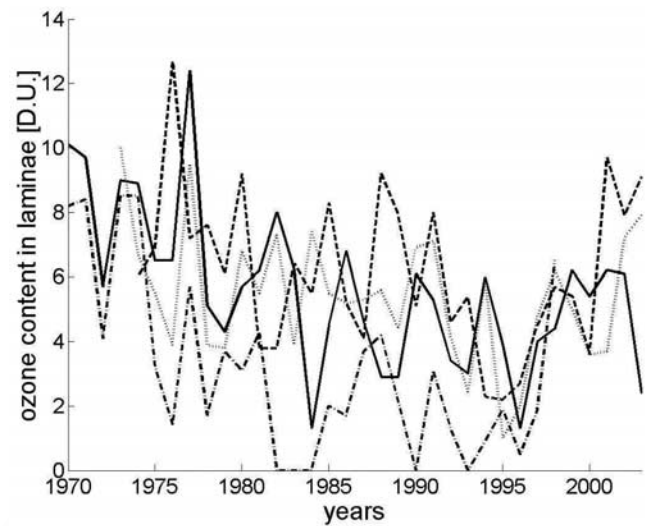


Figure 6. Overall ozone content in positive laminae per profile for the northern American middle latitude stations Churchill (dashed line), Edmonton (dotted line), Goose Bay (solid line), and Wallops Island (dash-dotted line), 1970–2003.

416 tions is small in the 1970s and 1980s, and the overall ozone
 417 content in laminae per profile is lower than that in Europe
 418 due to the lower latitudes of the Japanese stations. This
 419 influences the results and is also, but not only, responsible
 420 for the appearance of an outlier in Tateno in 1978. The
 421 ozone content in the laminae at Kagoshima is very low,
 422 because Kagoshima is the lowest-latitude station, which
 423 seems to be basically out of reach of the laminae of polar
 424 vortex edge origin. Nevertheless, we can say that there was
 425 again a substantial negative trend (Sapporo and Tateno)
 426 before the mid-1990 with a minimum around 1996, and an

evident tendency to trend reversal after this year, although 427
 not as rapid as in Europe. 428

[32] The Arctic stations (Figures 9 and 10) display a 429
 similar change in the trend of the overall ozone content in 430
 positive laminae as well as ozone deficit in negative laminae 431
 as the midlatitude stations with a minimum in 1995–1997. 432
 However, except for Resolute Bay all stations have provided 433
 data only since the very late 1980s. The negative trend 434
 followed by rapid trend reversal for positive laminae at 435
 Resolute Bay is similar to that for the above midlatitude 436
 stations. The overall ozone content in laminae per profile for 437
 high-latitude stations is higher than that for middle latitudes, 438
 which is in line with the idea of the dominant polar vortex 439

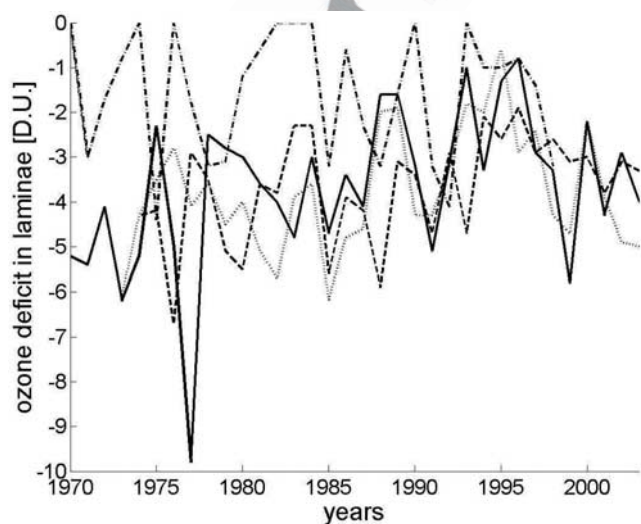


Figure 5. Overall ozone deficit in negative laminae per profile for the northern American middle latitude stations Churchill (dashed line), Edmonton (dotted line), Goose Bay (solid line), and Wallops Island (dash-dotted line), 1970–2003.

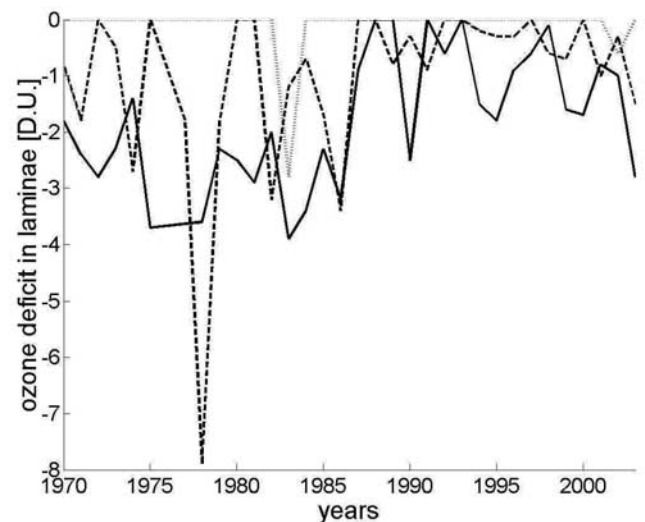


Figure 7. Overall ozone deficit in negative laminae per profile for the Japanese stations Kagoshima (dotted line), Sapporo (solid line), and Tateno (dashed line), 1970–2003.

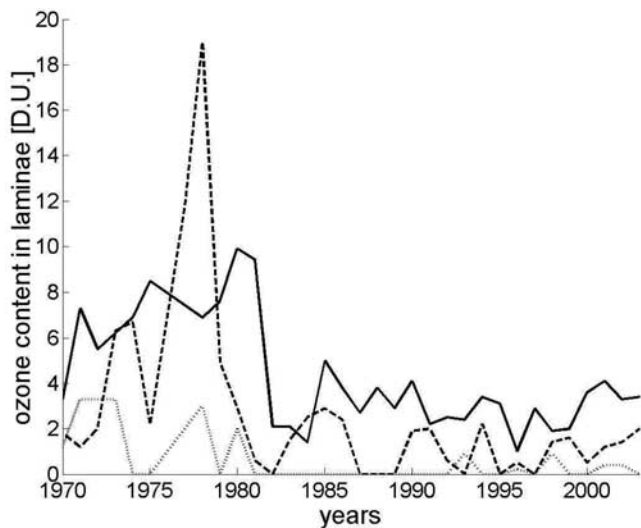


Figure 8. Overall ozone content in positive laminae per profile for the Japanese stations Kagoshima (dotted line), Sapporo (solid line), and Tateno (dashed line), 1970–2003.

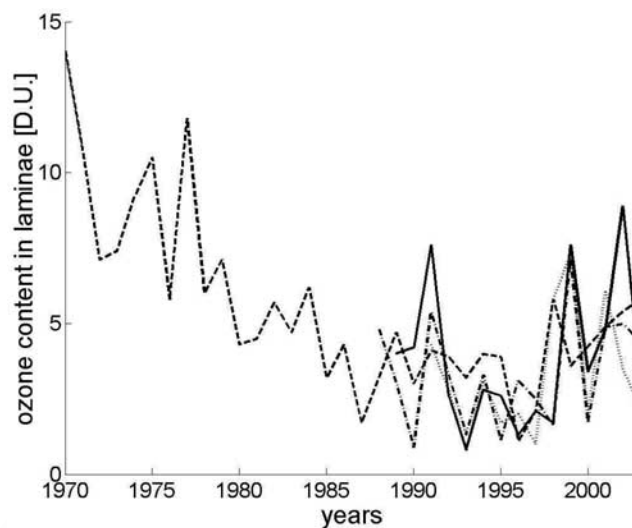


Figure 10. Overall ozone content in positive laminae per profile for the Arctic stations Alert (dash-dotted line), Ny Aalesund (dotted line), Resolute Bay (dashed line), and Sodankylä (solid line), 1970–2003.

440 edge origin of laminae. High-latitude stations are often
 441 situated inside the polar vortex. The ozone deficit to ozone
 442 surplus ratio seems to be around 0.6 for the Arctic stations.
 443 [33] The trend in the number of ozone laminae per profile
 444 is similar to that in the overall ozone content in laminae per
 445 profile, as shown in Figure 11 for the positive laminae and
 446 the representative stations of all four regions. The station
 447 with the largest amount of data, Payerne in Europe, reveals
 448 trends in terms of percentage change, which are very close
 449 to those in the overall ozone content in laminae per profile
 450 both for the positive and negative laminae. It is more
 451 difficult to make such a quantitative comparison of trends
 452 for other regions due to outliers and higher year-to-year
 453 variability, mainly because of the lower number of measure-

ments and their unstable seasonal distribution. Nevertheless, 454
 it is evident from the comparison of Figures 11 and 12 with 455
 Figures 4, 6, 8, and 10 that there is not much difference 456
 between the trends in the number of laminae per profile and 457
 the overall ozone content in laminae per profile for any of 458
 the regions under study. In Japan, Sapporo and Tateno 459
 display an evident trend in the number of laminae per 460
 profile. Kagoshima (not shown here) located at lower 461
 latitude than Sapporo and Tateno does not reveal a detect- 462
 able trend, but there are very few strong laminae at 463
 Kagoshima. The ratio of the number of negative laminae 464
 (not shown here) to positive laminae is approximately 2:3, 465

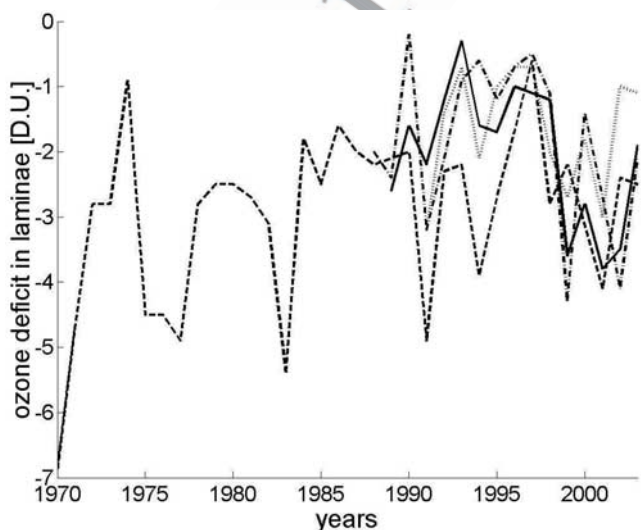


Figure 9. Overall ozone deficit in negative laminae per profile for the Arctic stations Alert (dash-dotted line), Ny Aalesund (dotted line), Resolute Bay (dashed line), and Sodankylä (solid line), 1970–2003.

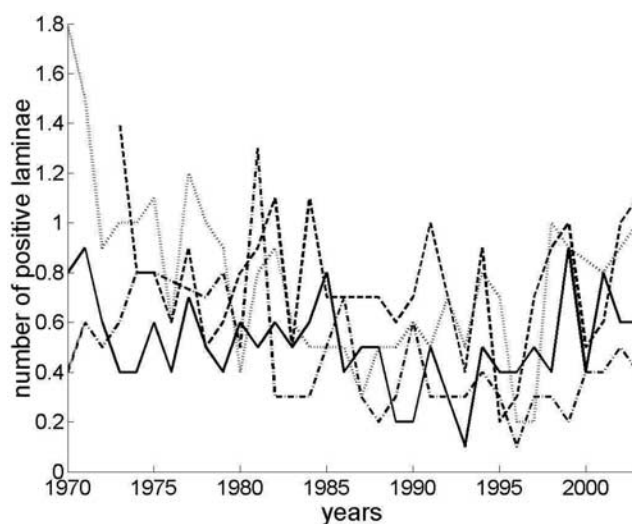


Figure 11. Number of positive laminae per profile for the four representative stations Payerne (Europe, solid line), Edmonton (northern America, dashed line), Sapporo (Japan, dash-dotted line), and Resolute Bay (Arctic, dotted line), 1970–2003.

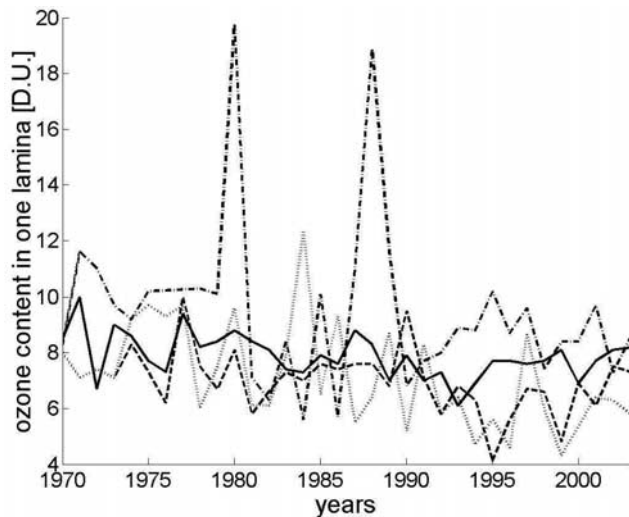


Figure 12. Ozone content per positive lamina for the four representative stations Payerne (Europe, solid line), Edmonton (northern America, dashed line), Sapporo (Japan, dash-dotted line), and Resolute Bay (Arctic, dotted line), 1970–2003.

466 i.e., similar to that for the overall ozone content in laminae
467 per profile.

468 [34] The third lamina parameter is the ozone content (or
469 deficit) per one positive (negative) lamina, shown for the
470 representative stations of all four regions in Figure 12 for
471 positive laminae. This parameter displays a quite different
472 pattern of trends. Payerne and Sapporo show a very weak
473 negative trend without detectable reversal, whereas the
474 middle and high-latitude stations in the American sector,
475 Edmonton and Resolute Bay, exhibit an evident negative
476 trend with a tendency to leveling off (Resolute) or even
477 slight reversal (Edmonton) of the trend in the mid-1990s.
478 On the other hand, there is no evident and statistically
479 significant trend for the negative laminae in any of the four
480 regions (not shown here). However, no detectable trend may
481 include the existence of a weak trend. Weak trends may be
482 masked by the problems with data uncertainty (instrumental
483 effects) mentioned at the end of section 3. In general, the
484 trends in the ozone content per lamina are less reliable than
485 trends in the other lamina parameters. The behavior of the
486 other stations in Europe and northern America, and Tateno
487 in Japan, is consistent with the behavior of the selected
488 representative stations Payerne, Edmonton and Sapporo.
489 The ozone deficit (not shown here) to ozone surplus
490 (positive laminae) ratio seems to be about 5:7, but its
491 estimate is very uncertain. The outliers for Sapporo occur
492 due to the smaller number of measurements and their year-
493 to-year unstable seasonal distribution.

494 5. Discussion

495 [35] The trends observed in the overall ozone content (or
496 deficit) in laminae per ozone profile are quite strong in all
497 four regions, Europe, northern America, Japan (middle
498 latitudes), and Arctic (high latitudes) down to about 35°N,
499 which means that the observed trend is a global character-
500 istic of the Northern Hemisphere at higher middle and high

latitudes. The decrease of the overall ozone content in
501 positive laminae per profile from 1970 to the mid-1990s
502 is more than 50%. The overall ozone content (deficit) in
503 laminae per profile is created by two parameters, by the
504 number of laminae per profile and the ozone content
505 (deficit) per lamina. In Europe and Japan, the trend is
506 almost entirely caused by the trend in the number of laminae
507 per profile. On the other hand, in the American sector this
508 only applies to the negative laminae, whereas for the
509 positive laminae the contribution of the trend in the ozone
510 content per lamina is significant, even though it is not
511 dominant. Such a difference in the origin of trends in the
512 American sector versus Europe and Japan was reported first
513 by *Mlch and Lastovicka* [1997] for positive laminae over
514 the period from the late 1960s to the early 1990s. They
515 found some difference in the trends in circulation indices in
516 the lower stratosphere, which could be at least partly
517 responsible for some difference in the origin of trends in
518 the overall ozone content in laminae per profile in the
519 American sector versus Europe and Japan.

520 [36] All stations in the Northern Hemisphere, except for
521 the lowest-latitude Kagoshima, show a reversal in the trend
522 in the overall ozone content in positive laminae per profile
523 in the mid-1990s. We assume that this reversal of the trend
524 is of dynamical origin, because it appeared too early for
525 chemical origin of the reversal (consequence of the Mon-
526 treal Protocol process). Such a reversal of trends has been
527 observed simultaneously in the total ozone content both in
528 satellite and ground-based (Arosa, Switzerland) data in the
529 middle latitudes of the Northern Hemisphere [*Appenzeller et*
530 *al.*, 2001; *Fioletov et al.*, 2002; *Fioletov*, 2004]; however,
531 no such trend reversal has been observed in the Southern
532 Hemisphere. There are also indications of a change of trends
533 in the zonal wind in the upper middle atmosphere in the
534 1990s [*Jacobi et al.*, 2003]. *Appenzeller et al.* [2001]
535 reported a tendency of the North Atlantic Oscillation
536 (NAO) from the 1970s to the mid-1990s to a more negative
537 phase, which was replaced by a tendency to a more positive
538 phase after 1995–1996. The ozone trends in the lower
539 stratosphere over Payerne at altitudes below 20 km were
540 found to be caused, to a substantial extent, by dynamical
541 changes [*Weiss et al.*, 2001]. All these observations suggest
542 rather a dynamical origin of the ozone laminae trend
543 reversal in the mid-1990s. There are various possible
544 dynamical contributors to the long-term trend in total ozone,
545 as discussed, e.g., by *Stahelin et al.* [2002] and *Hudson et*
546 *al.* [2003]. Our first multiple regression investigations for
547 Payerne, March average values, and a set of dynamical
548 solar parameters, reveal only four parameters, which are
549 responsible for more than 5% of the total variance each:
550 NAO (36%), potential vorticity (15%), QBO (at 50 hPa –
551 9%), Eliassen–Palm flux (5%). Any other parameter, includ-
552 ing El Niño–Southern Oscillation (ENSO), contributed less
553 than by 5%. However, for other regions we expect different
554 roles of dynamical parameters, for example for Japan the
555 role of ENSO must be larger and that of NAO smaller. A
556 more detailed investigation of the origin of the trend
557 reversal will be a topic of a separate paper after obtaining
558 information on laminae trends in the Southern Hemisphere,
559 which very likely establish some constraints on the possible
560 interpretation and origin/mechanism of the observed trend
561 reversal.

t2.1 **Table 2.** Coefficients of Correlation Between the Characteristics of Positive and Negative Laminae for All Ozone-sonde Stations Except Low-Latitude Kagoshima

t2.2	Stations	Number of Laminae	Ozone in Laminae per Profile	Ozone per One Lamina
t2.3	Payerne	0.68	-0.66	-0.29
t2.4	Hoheinpeissenberg	0.62	-0.79	0.08
t2.5	Lindenberg	0.64	-0.64	-0.11
t2.6	Legionowo	0.65	-0.48	-0.09
t2.7	Praha	0.84	-0.81	0
t2.8	Uccle	0.85	-0.83	-0.26
t2.9	Sodankyla	0.84	-0.80	-0.55
t2.10	Ny Aalesund	0.94	-0.80	-0.18
t2.11	Resolute Bay	0.51	-0.60	0.09
t2.12	Alert	0.80	-0.84	-0.34
t2.13	Goos Bay	0.76	-0.77	-0.29
t2.14	Edmonton	0.52	-0.63	-0.34
t2.15	Churchill	0.39	-0.65	0.26
t2.16	Wallops Island	0.33	-0.14	-0.29
t2.17	Sapporo	0.36	-0.46	0.13
t2.18	Tateno	0.85	-0.76	-0.09

563 [37] There might be some indications of a true “chemical” reversal, or rather leveling off of the trends in ozone in
 564 recent years since about 1997–1998, but in the upper
 565 stratosphere, as reported by *Newchurch et al.* [2003].
 566 However, *Steinbrecht et al.* [2004a] questioned the inter-
 567 pretation of the leveling off of the ozone trends in the upper
 568 stratosphere as evidence of “chemical” reversal. They
 569 attributed the observed changes rather to the solar cycle
 570 effect. The most recent paper by *Steinbrecht et al.* [2004b]
 571 mentions both possibilities and shows that measurements in
 572 the next few years (solar cycle minimum period) should
 573 resolve the problem.

574 [38] The results presented in section 4 indicate good
 575 correlations between the trends in positive and negative
 576 laminae parameters. Table 2 quantifies those correlations.
 577 We can see good correlation between the number of positive
 578 and negative laminae per profile; the correlation coefficient
 579 varies between 0.33 (Wallops Island) and 0.94 (Ny Aalesund).
 580 A good negative correlation is observed between the overall
 581 ozone in positive laminae per profile and the ozone deficit
 582 in negative laminae per profile, which is related to the good
 583 correlation between the number of positive and negative
 584 laminae. On the other hand, correlations between the ozone
 585 content per positive lamina and the ozone deficit per
 586 negative lamina are weak and mostly quite insignificant.
 587 This difference in correlations is understandable, if the main
 588 reason for correlations is the good correspondence between
 589 trends, because the trend observed in the ozone content per
 590 lamina is much smaller than in the other two parameters.
 591 However, some contribution to the observed correlations
 592 may be introduced by the method of lamina determination,
 593 namely by the smoothing used in constructing the reference
 594 ozone profile, which might result in a tendency to produce
 595 pairs of related positive and negative laminae therefore the
 596 results on the correlations between the positive and negative
 597 laminae characteristics must be considered to some degree as
 598 uncertain. Fortunately, as shown by *Krizan and Lastovicka*
 599 [2004] and mentioned in section 2, only the values of ozone
 600 content, not the trends in positive laminae characteristics are
 601 significantly influenced by the applied method of lamina
 602 determination.
 603

[39] *Lastovicka* [2002] estimated the contribution of the
 604 trends in laminae to the total columnar ozone trends at
 605 middle latitudes in Europe to be as much as one third of
 606 the overall trend in the late winter/early spring, and
 607 negligible in early autumn. However, this result was
 608 obtained with another method of lamina determination,
 609 which computed only positive laminae and with more
 610 ozone than the method we are using now. In other words,
 611 that estimate may be considered to be the upper limit of the
 612 effect of trends in laminae on trends in total ozone. The net
 613 ozone depletion caused by the difference between the
 614 overall ozone content versus deficit in laminae per profile
 615 (Figures 4 and 3) between 1970 and the mid-1990s is
 616 about 5–7% of the total ozone depletion for yearly average
 617 values. Since we use only January–May data, and with
 618 respect to the method of lamina determination used in this
 619 paper, which rather underestimates the laminae contribu-
 620 tion due to the connection between the positive and
 621 negative laminae, the estimate of 5–7% may be considered
 622 to be the lower limit of the effects of laminae on trends in
 623 total ozone in the late winter/early spring. Thus the
 624 contribution of laminae to the trends in total ozone is in
 625 no way dominant, but it cannot be neglected in European
 626 region, and similarly elsewhere at middle latitudes. The
 627 observation that the trends in total ozone at northern
 628 middle latitudes reveal a well-pronounced reversal in the
 629 mid-1990s in the late winter/early spring, but much weaker
 630 change in fall, is in line with the nonnegligible role of
 631 laminae in trends in total ozone.
 632

6. Conclusions

[40] The data of all the Northern Hemisphere ozone
 634 sounding stations at latitudes $\varphi > 30^\circ\text{N}$ with sufficiently
 635 long data series were analyzed for the long-term trends in
 636 laminae in ozone profiles in the lower stratosphere over the
 637 period 1970–2001. The analysis was complemented with
 638 several shorter data series, particularly from Arctic. The data
 639 were analyzed separately for four regions: Europe, northern
 640 America, Japan and the Arctic. As mentioned in section 1,
 641 the change of trends in laminae in the Northern Hemisphere
 642 was the main topic of the paper. Such a change in trends in
 643 the overall ozone content in positive laminae and the overall
 644 ozone deficit in negative laminae per profile was observed
 645 in the mid-1990s in the form of a trend reversal. The main
 646 results may be summarized as follows:
 647

[41] 1. There was a strong trend of reduction of the
 648 overall ozone content in positive laminae, as well as the
 649 overall ozone deficit in negative laminae per ozone profile
 650 and in the number of both positive and negative laminae per
 651 profile from 1970 to the mid-1990s in all four regions. The
 652 reduction reached more than 50% for the overall ozone
 653 content in positive laminae.
 654

[42] 2. After the mid-1990s the trend in both parameters
 655 and both positive and negative laminae reversed from
 656 negative to positive in all four regions.
 657

[43] 3. The trends in the number of laminae are very
 658 similar to those in the overall ozone content (or deficit) in
 659 positive (negative) laminae.
 660

[44] 4. There is no detectable trend in the ozone amount
 661 per positive lamina except for the American sector, where a
 662 weaker negative trend with a tendency to reversal or
 663

leveling off in the mid-1990s was observed, and no detectable trend in the ozone deficit per negative lamina.

[45] 5. There are some indications that the observed reversal of trends in laminae is of dynamical rather than chemical origin, i.e., unfortunately it probably is not a consequence of the Montreal Protocol and its amendments.

[46] Only large laminae, in terms of partial ozone pressure larger than 40 nbar, were analyzed. However, as mentioned in section 2, the trends appear to be almost independent of the size of laminae.

[47] Future work will be focused on two problems: (1) to obtain information about the trends in laminae in the Southern Hemisphere and (2) to find the origin of the observed reversal of laminae trends in the mid-1990s. Task 1 is a necessary condition for fulfilling task 2 because the behavior of trends in total ozone at middle latitudes of the Northern and Southern Hemisphere differs. Therefore the trends in laminae in the Southern Hemisphere can provide some constraints for the origin/mechanism of the trend reversal. To fulfill task 2, we have to find optimal proxies, mainly dynamical proxies, which influence the behavior of laminae. Such proxies must have global influence, because the reversal of trends in the ozone laminae characteristics has been observed in the whole Northern Hemisphere poleward of about 35°N. We shall focus our effort on the positive laminae behavior, because the negative laminae behave in a similar way as for the long-term trends.

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