

Effects of Energetic Particle Precipitation on the Mesosphere and Stratosphere

G.P. Stiller

IMK-ASF, Karlsruhe Institute of Technology

1 08.10.2009 2nd International HEPPA Workshop Boulder, CO, 6-8 October 2009



Forschungszentrum Karlsruhe n der Helmholtz-Gemeinschaft



Universität Karlsruhe (TH) Research University · founded 1825

Outline



Ionisation of the atmosphere by EPP

- Energy spectrum
- Penetration depth
- > Production of NO_x, HO_x, ions
- Ion models vs. parameterizations

Direct effects – SPEs

- Observations: MIPAS, ACE, GOMOS, HALOE, POAM, ODIN-SMR, SCIAMACHY, MLS
- NOx and ozone
- ➢ Ion (cluster) reactions: HO_x, Cl_x, HNO₃
- $> N_2O$
- > Model results: Jackman, Verronen, Winkler, Semeniuk

Indirect effects: EEPs

- Downward transport through dark mesosphere
- Dynamics vs. source strength
- Effects on stratospheric chemistry

Modeling of effects on ozone

- Short and medium term days to winters
- Long terms solar cycle





Sun-Earth connection





Turunen et al. 2009

2nd International HEPPA Workshop 08.10.2009 3 Boulder, CO, 6-8 October 2009

Forschungszentrum Karlsruhe in der Helmholtz-Gemeinscha



Universität Karlsruhe (TH) Research University + founded 1825

Energy deposition, penetration depth

- EPPs deposit their energy at different layers of the atmosphere depending on their penetration depth
- Energy required to create one ion pair is assumed to be 35 eV (Porter et al., 1976) or 36 ev (Rees et al., 1989)
- ⇒ Ionization rates estimated from energy deposition, various approaches



Formation of NO_x and HO_x from ion-electron pairs

NO_x: Dissociation of N₂ and O₂ to N₂⁺, O₂⁺, N⁺, O⁺, and NO⁺; formation of exited-state N(²D) and ground-state N(⁴S); $2N(^{2}D) + O_{2} \rightarrow 2NO$

HO_x: end of complex ion chemistry including protonated water cluster ions (up to 20 water molecules per proton)

Ion chemistry models calculate produced NO_x and HO_x ab initio (for example SIC, Verronen et al., Turunen et al.)

Parameterizations are also used: $2 HO_x$ molecules per ion pair in the USM,



energetic particles precipitate into atmosphere

Fig. 1. Particle precipitation effects on the ion-chemistry of the atmosphere.

<2 HO_x molecules per ion pair in the middle and upper mesosphere; 1.25 N atoms per ion pair, separated into 0.55 N(⁴S) and 0.7 N(²D) per ion pair (Jackman et al., 2008)

 NO_x and HO_x formation (plus further ion cluster formation) are the basis for all subsequent observed chemical changes in the stratosphere and mesosphere.







NO.

0₂(H₂O)₂

H₂O,M M O₂

0₂(H₂O)

м

H₂O

CIO

м

NC

M

NO3(HNO3) -HNO3.M м

NO₃(HNO

 CO_2

H₂O

 O_4^-

NO CO₂

03

0_{2.M}

CO,



0

N2 NO

6

Universität Karlsruhe (TH) Research University + founded 1825

Ozone depletion – catalytic cycles

 HO_x and NO_x react as catalytic substances in stratospheric ozone depletion cycles:

- $\begin{array}{l} \mathsf{OH} + \mathsf{O}_3 \to \mathsf{HO}_2 + \mathsf{O}_2 \\ \mathsf{HO}_2 + \mathsf{O} \to \mathsf{OH} + \mathsf{O}_2 \\ \underline{\mathsf{Net:}} \ \mathsf{O}_3 + \mathsf{O} \to \mathsf{2} \ \mathsf{O}_2 \end{array}$
- $\begin{array}{l} \mathsf{OH} + \mathsf{O}_3 \rightarrow \mathsf{HO}_2 + \mathsf{O}_2 \\ \mathsf{HO}_2 + \mathsf{O}_3 \rightarrow \mathsf{OH} + \mathsf{O}_2 + \mathsf{O}_2 \\ \underline{\mathsf{Net:}} \ 2 \ \mathsf{O}_3 \rightarrow 3 \ \mathsf{O}_2 \end{array}$

 $\begin{aligned} &\mathsf{NO} + \mathsf{O}_3 \to \mathsf{NO}_2 + \mathsf{O}_2 \\ &\mathsf{NO}_2 + \mathsf{O} \to \mathsf{NO} + \mathsf{O}_2 \\ &\underline{\mathsf{Net:}} \ \mathsf{O}_3 + \mathsf{O} \to \mathsf{2} \ \mathsf{O}_2 \end{aligned}$

 $\begin{array}{ll} \mathsf{ZO} + \mathsf{HO}_2 \to \mathsf{HOZ} + \mathsf{O}_2 & (\mathsf{Z:}\ \mathsf{CI},\ \mathsf{Br}) \\ \mathsf{HOZ} + hv \to & \mathsf{OH} + \mathsf{Z} \\ \mathsf{OH} + \mathsf{O}_3 \to \mathsf{HO}_2 + \mathsf{O}_2 \\ \mathsf{Z} + \mathsf{O}_3 \to \mathsf{ZO} + \mathsf{O}_2 \\ \underline{\mathsf{Net:}}\ \mathsf{2O}_3 \to \mathsf{3O}_2 \end{array}$

 $CI + O_3 \rightarrow CIO + O_2$ $CIO + O \rightarrow CI + O_2$ <u>Net:</u> $O_3 + O \rightarrow 2 O_2$

Etc. etc.

7 08.10.2009 2nd International HEPPA Workshop Boulder, CO, 6-8 October 2009







Boulder, CO, 6-8 October 2009



Direct effects – SPEs – NO_x and O₃

Why **direct** effect? Because effect is generated locally due to penetrating EPPs, without intermediate transport

Solar proton event Oct/Nov 2003: one of the largest events observed, many observations from a suite of satellite instruments available: MIPAS, SCIAMACHY, GOMOS, ODIN-SMR

Aftermath of the event in Arctic winter 2003/04: additional observations by ACE-FTS, HALOE, POAM

Solar proton event Jan 2005: observed by MIPAS, MLS, ACE-FTS, ODIN-SMR,...



Forschungszentrum Karlsruhe

in der Helmholtz-Gemeinschaft

Universität Karlsruhe (TH)

Research University • founded 1825

Other observations - SPE 2003 / 2005 – NO_x and O_3



http://atmos.caf.dlr.de/projects/scops/sciamachy_book/ sciamachy_book_figures/chapter_10/fig_10_32.jpg



Figure 4. (top) GOMOS daily (January 1–24, 2005) zonal mean night-time ozone mixing ratio [ppmv] at latitudes $65^{\circ}N-75^{\circ}N$. Note the destruction of the tertiary ozone maximum at 72 km altitude following the SPE on January 17, 2005. Contour lines as in Figure 3. (bottom) Ozone %-change (January 15–24) from the average of January 10–14. The contour lines are (-80, -70, -60, -50, -40, -30, -20, -10%). X-axis is the same as for the model results in Figure 2. Note the different x-axis in the two panels.

Seppälä et al., 2005

r Helmholtz-Gemeinschaft



Universität Karlsruhe (TH) Research University · founded 1825

Model results – SPE 2003 – NO_x and O_3



Research University + founded 1825



in der Helmholtz-Gemeinschaft

1108.10.20092nd International HEPPA WorkshopBoulder, CO, 6-8 October 2009

Model results – SPE 2003 – NO_x and O_3





Boulder, CO, 6-8 October 2009

12

Direct effects – SPEs – nitrogen, chlorine and hydrogen compounds

MIPAS observations during SPE Oct/Nov 2003 Further parameters from MIPAS available: Temperature, HNO₄, CH_4 , H_2O , CO, $BrONO_2$ (\Rightarrow poster Höpfner et al.), H_2O_2 (\Rightarrow poster Versick et al.).

MLS observations during SPE Jan 2005: OH and HO₂ (⇔ talk and poster Verronen et al.; poster Versick et al.)

HALOE observations during SPE July 2000: HCI (⇔ poster Winkler et al.)



in der Helmholtz-Gemeinschaft

Research University + founded 1825

13 08.10.2009 2nd International HEPPA Workshop Boulder, CO, 6-8 October 2009





Model results – SPE 2003 / 2005 – hydrogen compounds



OH during SPE Jan 2005 Case I: January 18, 12:10 LT Case II: January 20, 12:05 LT 90 90 Latitudes 69 - 70N Latitudes 70 - 72N 80 Longitudes 49W - 27E Longitudes 23W - 28E 80 Altitude [km] 20 20 Altitude [km] 00 02 40 40 30 30 6 n OH concentration [cm⁻³] OH concentration [cm⁻³] x 10⁶ $\times 10^{6}$ Case I : January 18, 21:20 LT Case II : January 20, 21:20 LT 90 90 80 80 Latitudes 71N - 72N Latitudes 71N – 72N Altitude [km] 20 20 Altitude [km] 09 02 Longitudes 35W - 41E Longitudes 22W - 29E 40 40 30 └─ _100 30└ -100 -50 50 100 -50 50 100 Relative change of ozone [%] Relative change of ozone [%]

O₃ during SPE Jan 2005 MLS observations, SIC (full ion chemistry) 1-D modelling Verronen et al., 2006



H₂O₂ during SPE Jan 2005 MIPAS observations, KASIMA 3D-CTM modelling

Forschungszentrum Karlsruhe

in der Helmholtz-Gemeinschaft

Versick et al., Poster session

Universität Karlsruhe (TH)

Research University + founded 1825

Summary direct (SPE) impact of EPP:

- High energetic protons penetrate deeply into the mesosphere and stratosphere and deposit their energy there
- \Box ~ 35 eV produce one ion-electron pair
- □ lons generate via more or less complicated ion (cluster) chemistry NO_x (mainly in the stratosphere) and HO_x (mainly in the mesosphere)
- □ NO_x and HO_x are involved in catalytic ozone destruction cycles ⇒ immediate ozone depletion observed and modeled
- □ Further perturbation of stratospheric/mesospheric chemistry is observed: HNO₃, N₂O₅, BrONO₂, HNO₄, N₂O, CO, CIONO₂, HCI, CIO, CIONO₂, OH, HO₂, H₂O₂,...
- Some of the chemical reactions can be modeled (by direct gas-phase or ion (cluster) chemistry), some are not yet understood.

Further topics to be discussed:

Impact of (secondary) electrons vs. protons
Effects on temperature and dynamics (e.g. via ozone depletion)
...





Indirect EPP effects

What are indirect EPP effects?





Auroral to high energetic electrons produce NO_x and HO_x in the upper mesosphere/lower thermosphere region (80 – 120 km).

The polar winter vortex acts as a "bathtub sink" where air is sucked down during polar winter.

But: The lifetime of HO_x is much too short to survive downward transport.

Only relevant species is NO_x . NO is produced in the MLT and can survive transport through the mesosphere in darkness only. Why?

18 08.10.2009 2nd Internat

09 2nd International HEPPA Workshop



NOx polar winter descent





NO_x in Antarctic polar winter 2003:



20

NO_x in Antarctic polar winter 2003: confinement to the dark vortex





HALOE observations

Energetic particle precipitation effects on the Southern Hemisphere stratosphere in 1992-2005

C. E. Randall,^{1,2} V. L. Harvey,¹ C. S. Singleton,¹ S. M. Bailey,³ P. F. Bernath,⁴ M. Codrescu,⁵ H. Nakajima,⁶ and J. M. Russell III⁷

D08308

RANDALL ET AL.: SH EPP EFFECTS 1992-2005



Figure 7. Maximum (black, solid) and average (dotted) excess NO_x densities in 2-week time periods from 15 May through 15 September, calculated as described in the text and Figure 6. Gray solid lines denote the number of molecules derived from the maximum excess densities (right axis, gigamoles). For clarity, the number of molecules in year 1994 has been allowed to run off-scale; numbers peak at 0.2 GM.

SH: Deposited NO_x in stratosphere reveals high interannual variability and correlates well with auroral activity and NO produced in the thermosphere

08.10.2009 2nd International HEPPA Workshop 22 Boulder, CO, 6-8 October 2009

Year-to-year **MAIT** variability of NO_x Subsidence in SH

D083

in de



Figure 10. EPP-NO_x calculated from the maximum NO_x residuals as in Figure 9 (dots, solid line, left axis) compared to auroral hemispheric power (top), medium energy electron hemispheric power (middle), and SNOE column NO from 97 to 150 km averaged over the sunlit region poleward of 60°S (bottom). Energetic particle and SNOE data (gray) are For averaged over the months of May-July and are referenced to the right vertical axes. Correlation coefficients are given in each panel.

High variability in NH



2nd International HEPPA Workshop

2003-2004 2004-2005 20ct Nov Dec Jan Feb Oct Nov Dec Jan Feb Oct Nov Dec Jan Feb OctNovDecJanFeb 60 Altitude [km] 50 12 10 8 40 9 30 60 5 Altitude [km] 50 40 2 .5 30 Oct Nov Dec Jan Feb GOMOS data Seppälä et al., 2007

High variability observed

High NO_x although geomagnetic activity was low: 2006, 2009

Unusually strong descent in 2004, 2006, 2009 NH stratospheric excess NO_x amounts seem to be ruled more by dynamics than by production See also talks by Randall and Funke

08.10.2009

23





Vivid discussion: Arctic winter 2003 - 2004



Figure 1. Combined observations of NO₂ during the Northern Hemisphere winter 2003–2004, showing (top) the >10 MeV proton flux (heavy line) and K_p index (light line), (middle) high-altitude ionization levels determined from the subionospheric radio wave index, and (bottom) GOMOS nightline and POAM III daytime NO₂ mixing ratios, with the POAM data shown inside heavy boxes. Both data sets have been zonally averaged over 2 d. Note the differing color scales for the two satellite data sets. These observations show the generation and descent of NO₂ into the upper stratosphere.

24

08.10.2009

Dynamic origin or in situ production?

Karlsruhe Institute of Technologic

- Correlation with dynamic tracers confirm downward transport of NO_x produced in the thermosphere (e.g. Funke et al., 2007; Hauchecorne et al., 2007)
- Semeniuk et al. (2005) found that SPE-generated thermospheric NO_x is not sufficient to explain amounts
 ⇒ additional production by auroral electrons
- Seppälä et al. (2007) found indication for contributions from aurorally generated NO_x

Seppälä et al., 2007





Impact of EEP-generated NO_x on stratospheric chemical budget: NO_v



1. Formation of upper stratospheric HNO₃



Boulder, CO, 6-8 October 2009

Solution Stiller et al., 2005) Solution Stiller et al., 2005)

- Every-year occurrence of upper strat. HNO₃ maximum confirmed by ODIN-SMR observations (Orsolini et al., 2009)
- Mechanism proposed by de Zafra and Smyshlyaev (2001):

 $\mathrm{N_2O_5}$ + H^+(H_2O)_n \rightarrow H^+(H_2O)_{n-1} + 2HNO_3 (only for z > 40km)

- N₂O₅ is formed immediately from NO_x under dark conditions
- Note: this is a different ion cluster reaction than during/after SPE!
- Mechanism confirmed by KASIMA model calculations (see Poster Reddmann et al.)
- 2. NO_x has impact on further NO_y species (e.g. BrONO₂, ⇔ poster Höpfner et al.)

Impact of EPP-generated NO_x on stratospheric chemical budget: O_3



Impact of EPP-generated NO_x on stratospheric chemical budget: Long-term O₃



WACCM3 without SPEs WACCM3 with SPEs

Both mean value minus average 1979-1980 * Observed ozone minus the observed average 1979-1980 NO_v production by SPEs

⇒Effect is very small

Long-term (decadal and longer) impact of EEP produced and descended NO_x ? Should be larger and more regular than by SPE-produced NO_x ! \Rightarrow Callis et al., 1998 covered 1979-1988





Summary: Indirect impact of EPP:



- \square NO_x and HO_x produced by auroral and high energetic electrons in the upper mesosphere and lower thermosphere
- Only NO_x is long-lived enough to be transported into the stratosphere by polar winter descent
- □ Transport through mesosphere possible only in darkness due to NO photolysis ⇒ confinement to dark polar winter vortex
- □ Import of NO_x during Antarctic winter highly variable and well correlated with geomagnetic activity ⇒ ruled by source strength
- □ Typical import amounts: 1 to 3 GMoles N per winter, this is about 5 to 15 % of the N_2O source.
- ❑ Arctic winters: extremely variable with exceptional winters 2004, 2006, 2009. High amounts also for years with low geomagnetic activity ⇒dynamical variability more important
- □ Winter 2003/2004: Discussion about descent vs. local production, contribution of SPE vs. EEP-generated NO_x
- Excess NO_x has impact on NO_y partitioning (upper stratospheric HNO₃ maximum produced by N₂O₅ conversion on protonated water ion clusters), BrONO₂, CIONO₂, ...
- \square Excess NO_x (and HO_x in the mesosphere) leads to short-term and medium-term (few months) ozone depletion
- □ Long-term ozone depletion by SPEs not significant
- □ Long-term ozone depletion by EEPs?







Certainly I have mentioned by far not all work regarding EPP impacts.

My apologies if any relevant observations, analyses, and modeling work are missing.

Thank you for your attention!

29 08.10.2009 2nd International HEPPA Workshop Boulder, CO, 6-8 October 2009



Forschungszentrum Karlsruhe n der Helmholtz-Gemeinschaft



Universität Karlsruhe (TH Research University - founded 182