

Baseline  
Observatory

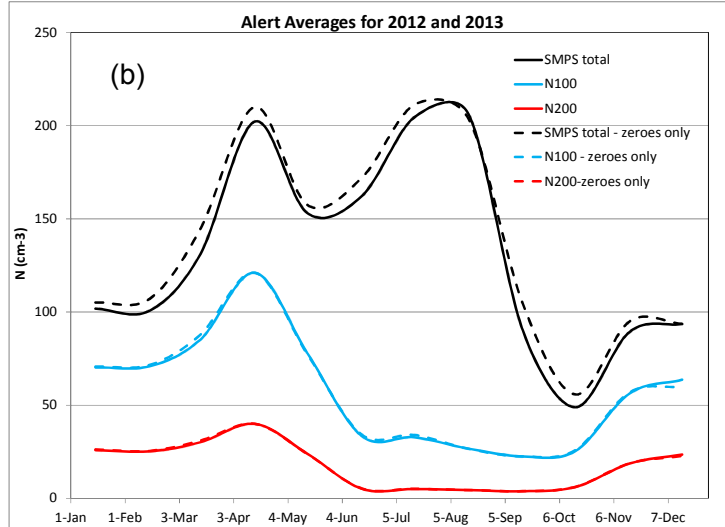


Figure S1. a) Location of Alert observatory relative to Alert station and air strip; b) monthly averages of number concentrations from SMPS with and without (zeroes only removed) the potential station influence removed (by wind sectoring and event removal). Variations in the volume estimated from the SMPS are represented by N200 (number concentrations of particles larger than 200 nm): N200 and volume correlate with a  $r^2$  of 0.997.

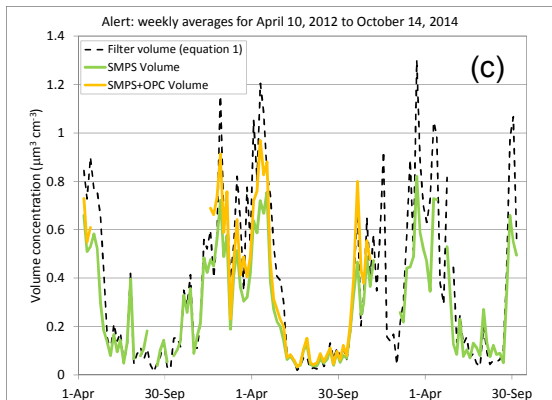
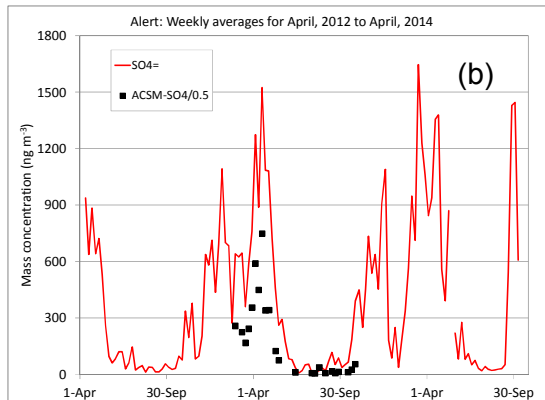
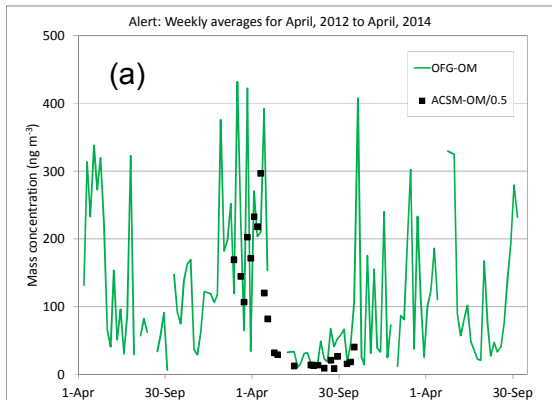


Figure S2. a) Time series of OM from the functional groups filter analysis (OFG-OM) and the ACSM (ACSM-OM); b) time series of sulphate from the IC filter analysis ( $\text{SO}_4^-$ ) and from the ACSM (ACSM-SO<sub>4</sub>); c) time series of volume concentrations estimated from the filters (equation 1), the SMPS (particles < 500 nm) and the SMPS+OPC (particles < 1  $\mu\text{m}$ ).

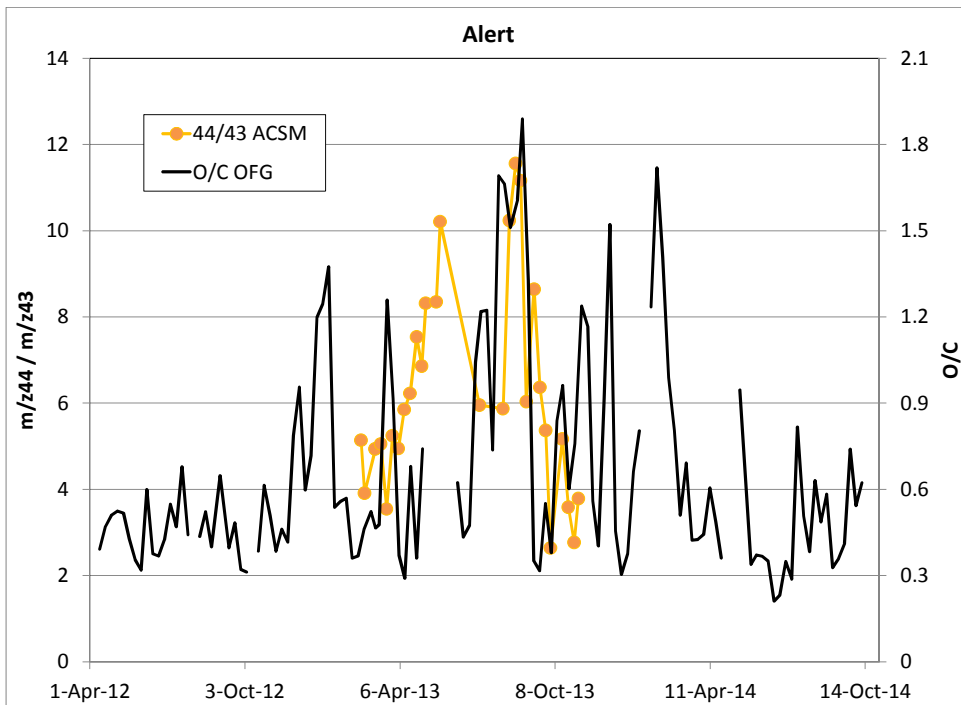


Figure S3. Time series of O/C calculated from the OFG as described in the text, and the ratio of m/z44 to m/z43 from the ACSM.

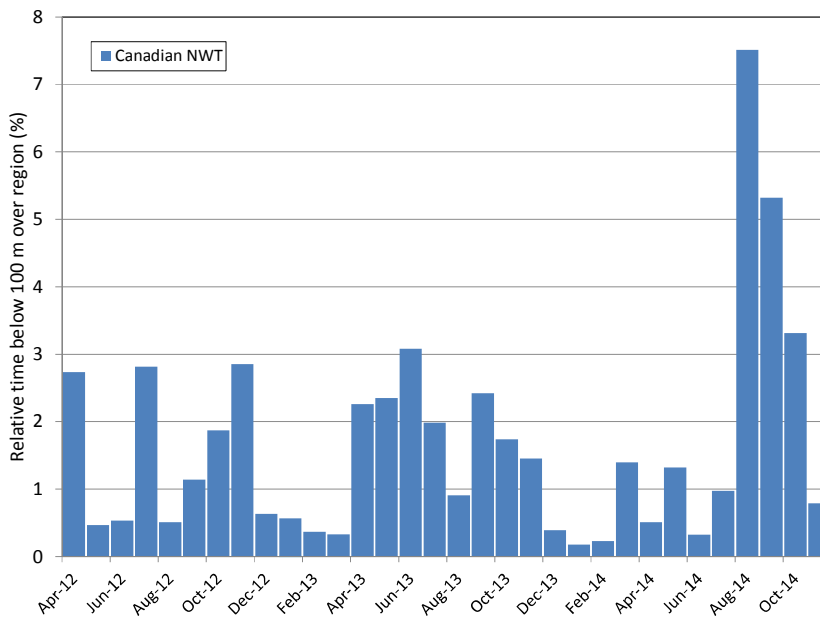


Figure S4. Monthly-averaged percentage of previous 10 days spent below 100 m over indicated regions that are identified in Fig. 1: a) regions 1-10; b) region 11 (Canadian NWT). All results from the FLEXPART trajectory analyses.

Table S1. Weekly-based coefficients of determination ( $r^2$ ) for particle mass concentration linear regressions with times spent over indicated regions. Values in bold indicate p of slope is  $<0.10$ . Values shaded gray indicate p of slope is  $<0.05$ . A minus sign in brackets indicates a negative slope. Uncertainties in the times over regions are  $<75\%$ .

Particle species	99 weeks with region 1	99 weeks with regions 1,4,6 and 7	30 dark <sup>#</sup> weeks with region 1	19 spring <sup>&amp;</sup> weeks with region 1
nss-SO <sub>4</sub> <sup>=</sup>	<b>0.07</b>	<b>0.05</b>	0.04	<b>0.19</b>
EC	<b>0.13</b>	<b>0.14</b>	0.08	0.12
ss-Na <sup>+</sup>	<b>0.18</b>	<b>0.16</b>	0.09	0.08
NO <sub>3</sub> <sup>-</sup>	<b>0.04</b>	<b>0.03</b>	0.00	0.01(-)
NH <sub>4</sub> <sup>+</sup>	<b>0.03</b>	0.03	0.05	0.00
nss-K <sup>+</sup>	<b>0.12</b>	<b>0.11</b>	0.05	0.20
nss-Mg <sup>++</sup>	<b>0.20</b>	<b>0.17</b>	<b>0.14</b>	0.15
nss-Ca <sup>++</sup>	<b>0.04</b>	<b>0.03</b>	0.00	<b>0.44</b>
MSA	<b>0.07(-)</b>	<b>0.08(-)</b>	0.00	0.05(-)
OM	0.01	0.01	<b>0.03</b>	0.05(-)
Alkane groups	0.01(-)	0.01(-)	0.00	0.13(-)
Alcohol groups	<b>0.05</b>	<b>0.03</b>	0.05	0.05(-)
Acid groups	0.00	0.00	0.02	0.12(-)
Amine groups	<b>0.12</b>	<b>0.10</b>	<b>0.15</b>	0.09

# - Weeks during all Novembers, Decembers, Januarys and Februarys.

& - Weeks during all March, April and May.

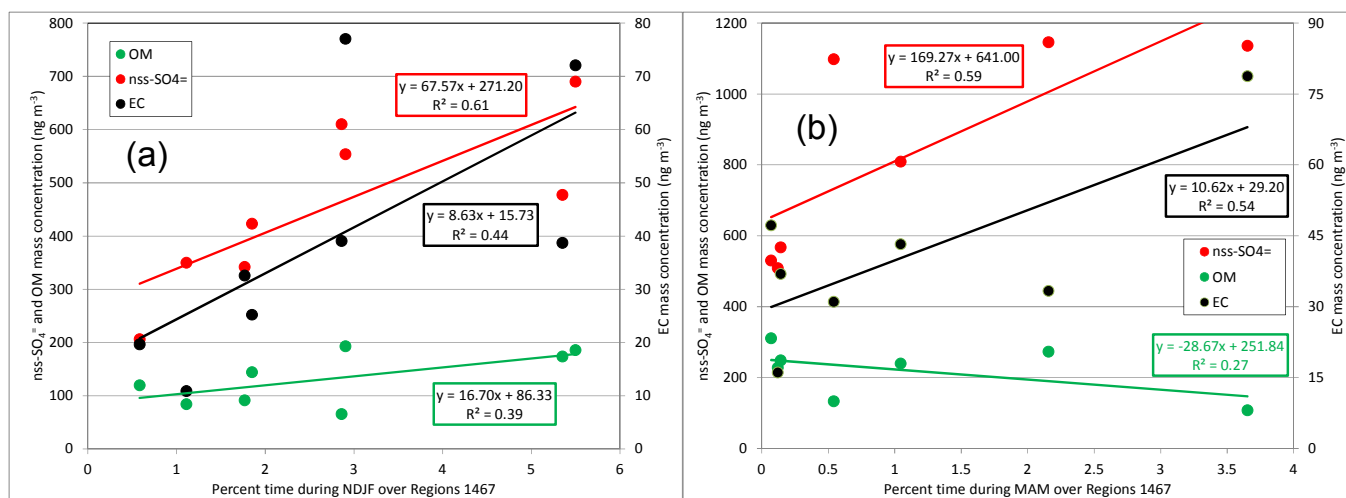


Figure S5. Plots of nss-SO<sub>4</sub><sup>=</sup>, EC and OM versus time spent over Regions 1467 (see text for definition of regions): a) dark months (NDJF); b) spring months (MAM).

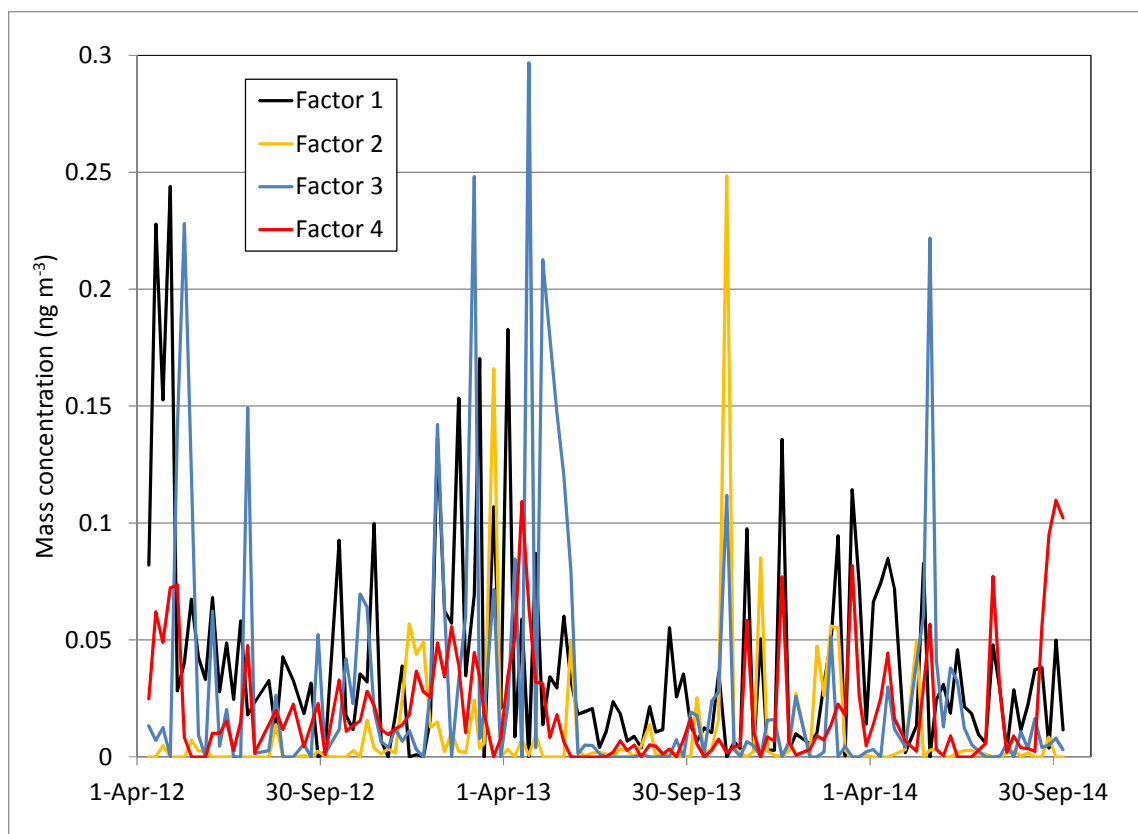


Figure S6. Time series of factor concentrations from PMF analysis.

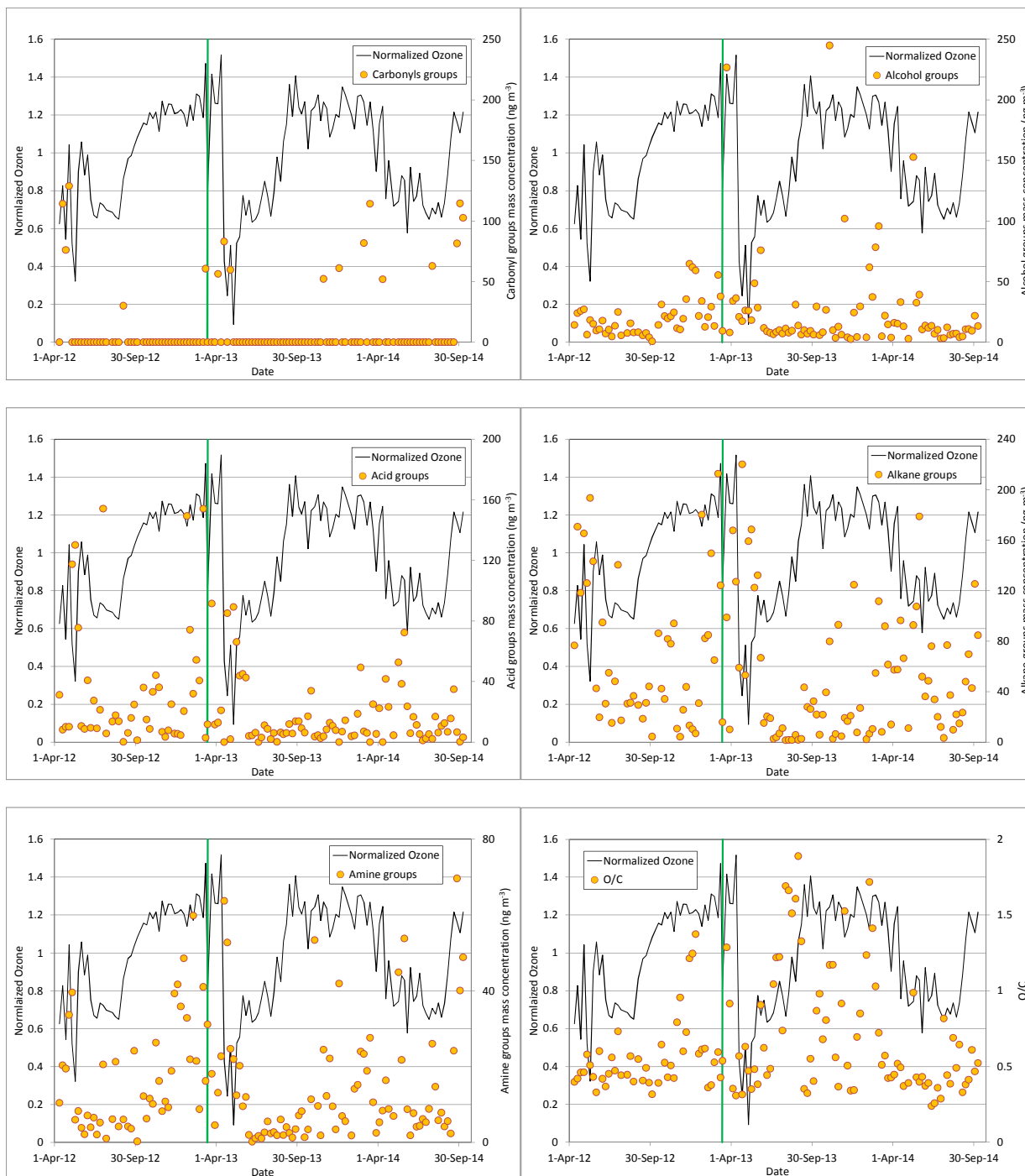


Figure S7. Time series of normalized ozone with carbonyl groups (a), alcohol groups (b), acid groups (c), alkane groups (d), amine groups (e) and O/C. All values are weekly averaged. The green line indicates time when problem with ozone inlet occurred (March 11, 2013). Ozone values before then are normalized to the average of values before March 11, 2013, and values after then are normalized to the average of values after March 11, 2013.



